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Important Exposure Factors for Children

An Analysis of Laboratory and Observational Field Data Characterizing Cumulative Exposure to Pesticides

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Important Exposure Factors for Children

An Analysis of Laboratory and Observational Field Data Characterizing Cumulative Exposure to Pesticides

By

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Notice

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Abstract

In an effort to facilitate more realistic risk assessments that take into account unique childhood vulnerabilities to environmental toxicants, the U.S. EPA's National Exposure Research Laboratory (NERL) developed a framework for systematically identifying and addressing the most important sources, routes, and pathways of children's exposure to pesticides. Four priority research areas were identified as representing critical data gaps in our understanding of environmental risks to children. Several targeted studies were conducted under NERL's children's exposure research program to specifically address these priority research needs. This document is a comprehensive summary report of data collected in these studies to address the priority research needs and is intended for an audience of exposure scientists, exposure modelers, and risk assessors. The parameters measured and the measurement methods are described. Data on representative organophosphate and pyrethroid pesticides are compared across studies and across compounds with the primary purpose of identifying or evaluating important factors influencing exposures along each relevant pathway. Summary statistics, comparative analyses, and spatial and temporal patterns are presented to address previously identified data gaps. Results are compared across studies in order to identify trends that might provide a better understanding of the factors affecting children's exposures. While highlights of the results of individual studies are presented, the focus is on presenting insights gleaned from the analysis of the aggregated data from several studies. By examining relationships among application patterns, exposures, and biomarkers for multiple compounds from different classes of pesticides, this report strives to help produce more reliable approaches for assessing cumulative exposure.

Executive Summary

In an effort to facilitate more realistic risk assessments that take into account unique childhood vulnerabilities to environmental toxicants, the National Exposure Research Laboratory (NERL) in the U.S. Environmental Protection Agency's (U.S. EPA) Office of Research and Development (ORD) developed a framework for systematically identifying and addressing the most important sources, routes, and pathways of children's exposure to pesticides (Cohen Hubal *et al.,* 2000a, 2000b). Using this framework, a screening-level assessment was performed to identify the exposure pathways with the greatest potential exposures. The uncertainty associated with assessing exposure along each pathway was then evaluated through an exhaustive review of available data. Four priority research areas were identified as representing critical data gaps in our understanding of environmental risks to children. The absence of sufficient real-world data in all four of these areas produces an excessive reliance on default assumptions when assessing exposure. These *priority research areas* are: 1) pesticide use patterns; 2) spatial and temporal distributions of residues in residential dwellings; 3) dermal absorption and indirect (non-dietary) ingestion; and 4) dietary ingestion.

Several targeted studies were conducted or financially supported by NERL under the children's exposure research program to specifically address these priority research needs. These studies included:

- Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants ("CTEPP")
- First National Environmental Health Survey of Child Care Centers ("CCC")
- Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposures in Children Living in Jacksonville, Florida ("JAX")
- Center for the Health Assessment of Mothers and Children of Salinas Quantitative Exposure Assessment Study ("CHAMACOS")
- Children's Pesticide Post-Application Exposure Study ("CPPAES")
- Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the US EPA Indoor Air Quality Test Research House ("Test House")
- Pilot Study Examining Translocation Pathways Following a Granular Application of Diazinon to Residential Lawns ("PET")
- Dietary Intake of Young Children ("DIYC")
- Characterizing Pesticide Residue Transfer Efficiencies ("Transfer")
- Food Transfer Studies ("Food")
- Feasibility of Macroactivity Approach to Assess Dermal Exposure ("Daycare")

Two studies performed prior to the identification of priority research areas also provided useful data. These were:

- National Human Exposure Assessment Survey in Arizona (NHEXAS-AZ)
- Minnesota Children's Pesticide Exposure Study ("MNCPES")

All studies involving children were observational research studies, as defined in 40 CFR Part 26.402. All study protocols and procedures to obtain the assent of the children and informed consent of their parents or guardians were reviewed and approved by an independent institutional review board (IRB) and complied with all applicable requirements of the Common Rule regarding additional protections for children. Further, all protocols regarding recruitment and treatment of participants were reviewed by the EPA Human Subjects Research Review Official (HSRRO) to assure compliance with the Federal Policy for the Protection of Human Subjects.

The studies took place in EPA research laboratories, in the EPA Indoor Air Quality Research Test House, in private residences, and in child care centers. The studies have been grouped as a) large observational field studies (NHEXAS-AZ, MNCPES, CTEPP, and CCC), b) small pilot-scale observational studies (JAX, CPPAES, DIYC, CHAMACOS, and Daycare), and c) laboratory studies (Test House, Transfer, and Food). The large observational field studies had either a regional (NHEXAS-AZ, MNCPES, CTEPP) or national (CCC) focus. A broad suite of chemical contaminants, including organophosphate and pyrethroid pesticides and their metabolites, were typically measured in multiple environmental media and in urine. Some of the small pilot-scale studies included measurements of multiple chemicals in multiple media in locations either with year-round residential pesticide use (JAX) or in close proximity to agricultural fields (CHAMACOS). Other pilot-scale studies focused on a single compound (CPPAES, DIYC, PET, Daycare). The laboratory studies (Transfer, Food, Test House) evaluated factors affecting transfer from surfaces or investigated post-application spatial and temporal variability. One of the primary objectives for all of these studies was to determine and quantify the key factors that influence exposure along the pathways relevant to the four priority research areas.

This document is a comprehensive summary report of data collected under the NERL children's exposure research program and is intended for an audience of exposure scientists, exposure modelers, and risk assessors. The parameters measured and the measurement methods are described. Data on representative organophosphate and pyrethroid pesticides are compared across studies and across compounds with the primary purpose of identifying or evaluating important factors influencing exposures along each relevant pathway. Summary statistics, comparative analyses, and spatial and temporal patterns are presented to address previously identified data gaps. Results are compared across studies in order to identify trends that might provide a better understanding of the factors affecting children's exposures. While highlights of the results of individual studies are presented, the focus is on presenting insights gleaned from the analysis of the aggregated data from several studies. By examining relationships among application patterns, exposures, and biomarkers for multiple compounds from different classes of pesticides, this report strives to help produce more reliable approaches for assessing cumulative exposure.

With limited data available to EPA researchers on the types, locations, and frequency of pesticide usage in residential and other non-occupational environments, pesticide use patterns were identified as a priority research area. Accordingly, pesticide use information was collected by inventory and questionnaire in each of the field studies. Questionnaire items and inventory

forms differed, geographic regions represented were limited, and the total number of study participants was relatively small. Furthermore, during the period of four years covered (1997 to 2001), pesticide manufacturers were increasingly replacing organophosphates with pyrethroids in their formulations, and restrictions on residential applications of the most commonly used organophosphates were approaching. Nevertheless, important usage information was produced by the studies. Pyrethrins and their synthetic analogs (pyrethroids), specifically permethrin, cypermethrin, and allethrin, are clearly the most frequently used insecticides for indoor applications in homes and child care centers based on inventories and records. Organophosphates appear to persist in indoor environments, as chlorpyrifos and diazinon were more frequently detected in screening wipes (at frequencies comparable to permethrin) than in inventories. Among the carbamates, only propoxur and carbaryl were inventoried or reportedly used.

"Crack-and-crevice" type applications were used more often than either broadcast or total release aerosol ("fogger") applications. Applications were more likely to be performed by the resident than by a professional service in JAX, and also as reported in NHANES. In JAX, the modes of application included hand pump sprayer (37%), aerosol can (24%), fogger (3%), and baits (3%), but the pertinence of these results to other locations is unknown. Apart from these results, information on application type and method was not collected.

Pesticide products were found in at least 86% of JAX and MNCPES screening households, with a mean of three products per household. There is evidence in support of a pattern of higher application frequencies in warmer climates, with the percentage of participants reporting use in a given time period highest in Florida, lower in North Carolina and Ohio, and lowest in Minnesota. The percentage in Jacksonville, FL is substantially higher, and the percentage in Minnesota is substantially lower, than the national average reported in NHANES. In childcare centers, monthly interior pesticide applications were performed in about a third of the CCC facilities nationwide and were anecdotally found to be standard practice among daycares contacted in North Carolina.

There were no statistically significant differences in the total number of products found or reportedly used in MNCPES based on either population density (urban vs. non-urban households) or other socio-demographic factors including race, ethnicity, home type, income, and level of education. Similarly, analysis of CTEPP data found no association between application frequency and either population density or income class.

A second primary research area is spatial and temporal distributions of pesticides in residential dwellings. Spatial and temporal heterogeneity may affect exposure estimates along all exposure routes. Absorption via the inhalation route relies on the measured airborne concentration. Absorption via the dermal and indirect ingestion routes relies on the measured surface loading. Even estimates of dietary ingestion for children may depend on surface concentrations due to pesticide transfer during food preparation and handling. Examination of distribution patterns of airborne and surface residues has yielded important insights.

The organophosphate insecticides chlorpyrifos and diazinon were most frequently detected in both indoor air and outdoor air in these field studies, but the detection frequencies in outdoor air were lower and more variable across studies. Chlorpyrifos was frequently detected even after its

indoor residential use was restricted, perhaps due to emissions from indoor sinks (*e.g.*, carpets) and from continued use of existing home inventories. Indoor air concentrations were typically an order of magnitude higher than outdoor air concentrations, with notable exceptions of outdoor diazinon and permethrin levels which were nearly as high as indoor levels in JAX, and outdoor diazinon levels that exceeded indoor levels in the agricultural community monitored in CHAMACOS. The low pesticide concentrations routinely measured outdoors (notwithstanding the exceptions noted) together with the relatively short time spent outdoors suggests that inhalation of outdoor air is not typically an important contributor to aggregate pesticide exposure. The similarity across large observational field studies in the variability of the observed indoor air chlorpyrifos concentrations, despite sample collection periods ranging from 1 to 7 days, suggests that air concentrations are reasonably consistent from day-to-day in the absence of a recent application.

The median indoor air concentrations of the organophosphates are higher than that of the pyrethroids. While these studies were conducted at a time when organophosphates arguably dominated the marketplace, a comparison of the mean levels of various organochlorine, organophosphate, and pyrethroid pesticides measured in CTEPP finds that the concentrations measured in the absence of recent applications appear to be strongly influenced by vapor pressure, with the more volatile pesticides, such as chlorpyrifos, found at the highest levels. Consequently, the importance of inhalation as a route of exposure for pesticides is likely to decrease as less volatile pesticides, such as the pyrethroids, are introduced into the market.

Differences in sampling methods, year of the study, and time of year when samples were collected make it difficult to distinguish any regional differences in pesticide concentrations. In general, median indoor air concentrations were somewhat higher in southern states (NHEXAS-AZ and CTEPP-NC) than in northern states (MNCPES and CTEPP-OH). However, the distributions exhibit considerable overlap across geographical locations. When daycare measurements are included, a geographical difference is less obvious, perhaps due to regular, calendar-based pesticide treatments at many daycare facilities.

Irrespective of region, differences in indoor air levels between homes and daycares were not found to be statistically significant. Similar mean indoor air levels observed in homes and daycares demonstrate the potential for continued exposure as a child spends time in other indoor locations. Additional concentration measurements in other locations would be useful to examine exposure potential from different settings such as schools, restaurants, and other public and private locations where pesticides are also applied.

Differences in indoor air concentrations associated with population density and income level were observed in the field studies. Differences between urban and rural air concentrations were observed in both MNCPES and CTEPP. In fact, urban chlorpyrifos levels were about 25% higher than rural levels across studies. A reasonable explanation may be that urban areas require more intensive use of pesticide products to control a range of pests over a wider seasonal span. Concentrations of chlorpyrifos and diazinon were higher in low-income homes than in medium/high income homes in CTEPP, but the difference was statistically significant only for diazinon, and only in NC.

Within-home spatial and temporal patterns were investigated following a crack and crevice application of chlorpyrifos in the kitchen of the Test House. The pesticide was detected even in the farthest bedroom from the application, with a concentration gradient observed from the kitchen to the den (proximal area) to the master bedroom (distal area). Temporally, airborne concentrations peaked on day 1, then decreased by approximately 80%, but were still measurable, at 21 days after application. In contrast, airborne diazinon concentrations among homes in the DIYC study were most pronounced 4-5 days after application. Between-home spatial variability following a pesticide application was investigated in the CPPAES study. Indoor air chlorpyrifos concentrations spanned more than an order of magnitude among the homes one day after application.

Significant progress has also been made in understanding spatial and temporal distributions of organophosphate residues on surfaces. In a published analysis of the MNCPES surface wipe data, Lioy and colleagues (2000) reported substantial variability in surface chlorpyrifos levels among different rooms. Substantial variability among and within rooms is also evident in the Daycare data. Furthermore, data from the Test House also show that surface loadings cannot be assumed to be homogenous even within a room. These observations suggest that multiple locations should be sampled to more accurately represent surface loadings. Exposure modelers using probabilistic methods have already begun to account for differences in surface loadings based on proximity to application sites in order to reduce possible exposure misclassification in their exposure estimates.

A number of observations suggest that there is substantial translocation of pesticides from application surfaces to adjacent surfaces, but levels remain higher at the application location. In CPPAES, the post-application chlorpyrifos loadings were higher than the pre-application values even on surfaces that did not receive a direct application. In DIYC, the transferable residues on the counters were nearly as high as those on the floors immediately after application. In JAX, the application area surface residue loadings were generally higher than the play area surface residue concentrations. In the CCC, the floor residue loadings were generally higher than the desk top loadings. High loadings of diazinon in indoor house dust following the lawn treatment in the PET study suggest that transfer into the house may also occur.

Examination of chlorpyrifos and diazinon loadings following applications indicates that *total available residue* loadings decay at a slower rate than airborne concentrations. Total available residue loadings (obtained by methods intended to measure the total amount of contaminant on a surface) also appear to decline at a slower rate than *transferable residue* loadings (intended to represent the amount that is transferred as a result of contact with the contaminated surface). In fact, using a total available residue method, chlorpyrifos was measured in 62% of the MNCPES samples, even in the absence of a recent pesticide application.

On a regional level, Jacksonville, Florida, an area known for year-round pest control issues and identified as having high pesticide usage during the NOPES study (Whitmore *et al.,* 1994), had much higher surface concentrations than any of the other studies without recent applications. Within a given region, however, there appears to be little relationship between questionnaire information and measured surface values. Previously published results from the MNCPES indicate that the residential pesticide use questions and overall screening approach used in the MNCPES were ineffective for identifying households with higher levels of individual target

pesticides (Sexton *et al.*, 2003). Results from the CPPAES study suggest that cleaning activities and ventilation influence surface concentrations; it appears that the surface chlorpyrifos loadings were lower in those homes in which the occupants reported additional cleaning activities and/or high ventilation rates.

While significant progress has been made in understanding spatial distributions of organophosphate and pyrethroid pesticides in the absence of a recent application and in understanding spatial and temporal distributions of organophosphate pesticides following an application, no data have been produced on the spatial and temporal distributions of pyrethroids following applications. The movement of residentially applied insecticides follows a complex and poorly understood process of transformation and phase distribution and is influenced by several factors. Differences in physicochemical characteristics make it difficult to generalize the spatial and temporal distributions of organophosphate pesticides to pyrethroid pesticides, but with information on chemical properties and on human activities, distribution patterns can be modeled.

The third primary research area was identified as dermal absorption and indirect ingestion. Intake via these exposure routes is often estimated using measurements of pesticide concentrations in dust and soil and pesticide loadings on surfaces. Intake estimates also rely on numerous default exposure factor assumptions. Pesticides in dust generally had high detection frequencies, consistent with dust being considered a repository of contaminants. Detection frequencies for soil samples, on the other hand, were generally low (with the exception of measurements made immediately following lawn applications).

Compounds found at relatively higher concentrations in dust tend to be found at relatively lower concentrations in air. The less volatile pyrethroid pesticides tend to partition to the dust and may degrade more slowly allowing accumulation over time from repeated applications. This underscores the importance of dust as a primary residential exposure medium for the less volatile pesticides. In addition, the exposure factors that are important for other nonvolatile contaminants such as lead may also be important for the less volatile pesticides.

Pyrethroids generally have low vapor pressures and Henry's Law constants, thus they are poorly volatilized and exist almost entirely in the particulate phase at room temperature. Furthermore, high octanol/water (K_{ow}) and water/organic carbon (K_{oc}) partition coefficients cause pyrethroids to partition into lipids and into organic matter. With these characteristics, pyrethroids can be expected to bind readily to the particulate matter that comprises house dust. Particles resuspended by human activity then act as the primary vector for pyrethroid transport and for human exposure. Particle-bound movement and transfer of pyrethroids imply a decreased importance of the inhalation route and an increased importance of routes that involve dermal transfer, such as indirect ingestion and dermal absorption. Exposure of young children, for whom indirect ingestion of residues from object- and hand-to-mouth activities is particularly important, may be most strongly affected. In fact, algorithm-based estimates of distributions of intake of chlorpyrifos and permethrin from the four contributing routes among the CTEPP-OH children indicated that the contribution from the indirect route is much more important for permethrin than for chlorpyrifos.

Comparisons of pesticide surface loadings (ng/cm^2) showed higher levels in the CTEPP daycare centers than in the homes. This appears to be the result of higher amounts of dust in the daycare centers, as there is not as large of a difference in the pesticide concentrations (ng/g) in the dust. Studies with lead have suggested that loading may have a greater impact than concentration on actual intake, thus higher amounts of dust may be important even if the concentration within the dust is similar.

Data from our studies show that the collection methods utilized may have sizeable effects on estimates of dermal exposure and indirect ingestion. Total residue methods, which use both solvent and mechanical action to remove residues that may have penetrated into the surface, produce the highest values, followed by dust methods, and then by transferable residue methods. These methods are intended to measures different types of transfer, and efficiencies for various methods have been previously published. Use of total residue methods allows the assessor to use appropriate transfer factors to represent a transfer efficiency applicable to a given scenario. Questions remain, however, on exactly how much of what is measured by total residue methods is truly available for transfer and how much would otherwise be trapped in the pores and/or body material of the surfaces if not for the mechanical and solvent action of the methods.

Even the amount of solvent used with wipe samples affects the results. The low pesticide surface loadings obtained with 2 mL isopropyl alcohol wipes in both the NC and OH CTEPP studies (loadings similar to those obtained with the polyurethane foam [PUF] roller) suggest that the amount of IPA applied to the wipe may affect the amount of pesticide residue recovered. Surface type has also been shown to affect the collection efficiency of wipes. Recently published NERL data (Rohrer *et al.,* 2003) found that with respect to pesticide transfer, wiping from hard surfaces greatly exceeded carpet, and wiping from tile generally exceeded hardwood. Clearly, some standardization of surface sampling methods is needed.

Although successfully used in laboratory studies, the Modified C18 Surface Press Sampler was rarely able to measure pesticide residues in field studies. The original press sampler was designed to measure transfer of dust-bound pesticides to the skin from a single hand press onto a carpeted surface. The uses for the modified C18 surface press sampler have expanded to include hard surfaces and longer contact times, effectively using the press sampler in a manner for which it was not intended. Our data suggest that the sensitivity of the modified C18 surface press sampler may be too low to measure residential pesticide residues (which may transfer by both equilibrium mass transfer and mechanical transfer).

Laboratory studies using fluorescent tracers (as surrogates for pesticide residues) indicated that *tracer type*, *surface type*, *contact motion*, and *skin condition* were all significant factors. Transfer was greater with laminate (over carpet), smudge (over press), and sticky skin (over moist or dry). *Contact duration* and *pressure* (force) were not found to be important factors. The effect of surface type appeared to diminish with repeated contact, while the effect of skin condition (moist vs. dry) appeared to increase with repeated contact. Additional studies are still needed to gain a better understanding of the key factors that influence the dermal transfer and indirect ingestion of pesticides.

The frequencies of hand- and object-to-mouth contacts were quantified for preschool children in the CTEPP and CPPEAS studies using the Virtual Timing Device (VTD) software (Zartarian *et al.*. 1997). The CPPAES results support the use of the commonly assumed median count of 9.5 hand-to-mouth contacts per hour; however CTEPP data suggest a much higher value for younger children. The CTEPP methodology also accounts for combination hand- and object-to-mouth contacts during both eating and non-eating events.

The fourth primary research area was identified as dietary ingestion. Diet can be an important pathway of exposure. Foods may contain residues of pesticides and other environmental chemicals because of intentional applications or may become contaminated during processing, distribution, storage, and consumption. For certain chemicals, diet is potentially the *predominant* pathway of exposure. Children's dietary exposure to pesticides is not limited to the residues in or on foods when they are brought into the home. Children's unique handling of foods prior to consumption requires special attention, but it is rarely considered in study designs.

Based on route-specific intake estimates, dietary ingestion represented the dominant route of exposure for chlorpyrifos, diazinon, and permethrin in the CTEPP study. Unfortunately, the route that represented the dominant route of exposure was also the route with the lowest detection frequencies (approximately 2/3 of the values for permethrin in CTEPP were nondetects), which increases the uncertainty in the estimates. Substituting a fraction of the detection limit for values below the limit of detection may have a disproportionate impact on assessing the importance of the dietary route.

The most common measure of dietary exposure was by composited duplicate diet analyses. However, great care must be taken to ensure that the duplicate diet accurately reflects what is actually consumed instead of what is served because significant quantities of food may remain uneaten by children. Duplicate diets fail to capture those pesticide residues transferred to foods as a result of the child's handling of food prior to and during consumption. In DIYC, estimates of dietary intake that included excess contamination due to handling were as much as double the estimates of intake based on duplicate diet alone. These results suggest that dietary estimates based on duplicate diet may not be as reliable for young children as they are for adults.

Progress has been made in many areas and we are beginning to understand the environment that children live in, their activities, and the resulting exposures. However, research is still needed to adequately characterize the magnitude, routes and pathways of exposure. We still need to understand the key factors that influence the dermal transfer and indirect ingestion of pesticides. We need to be able to more accurately assess dietary exposure. In order to evaluate exposure models, we must be able to quantify the relationships between and among environmental concentrations of pesticides in various media, children's activities, and the results of biomarkers of exposures as measured in urine and/or blood. Exposure models outputs that include the timing and route of exposure need to be linked to PBPK models in order to develop accurate assessment of target tissue dose. Research, especially model development, needs to extend beyond single chemical aggregate exposures and dose to include exposures and risks that accumulate across chemicals and over time.

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Abbreviations and Acronyms

%Det	Percent of samples above detection limit
2,4-D	2,4-Dichlorophenoxyacetic acid
3-PBA	3-Phenoxybenzoic acid
ACH	Air exchanges per hour
AER	Air exchange rate
AEV	Application effective volume
ANOVA	Analysis of variance
ASTM	American Society for Testing and Materials
ATSDR	Agency for Toxic Substances and Disease Registry
AZ	National Human Exposure Assessment Survey in Arizona
C18 Press	C18 surface press sampler
CCC	First National Environmental Health Survey of Child Care Centers Study
CDC	Centers for Disease Control
CDIM	Children's Dietary Intake Model
CHA	Center for the Health Assessment of Mothers and Children of Salinas Quantitative Exposure Assessment Study
CHAMACOS	Center for the Health Assessment of Mothers and Children of Salinas Quantitative Exposure Assessment Study
cis-P	*cis*-Permethrin
c-Perm	*cis*-Permethrin
c-Permethrin	*cis*-Permethrin
CPPAES Pre	CPPAES Study, pre-application days only
CPPAES	Children's Pesticide Post-Application Exposure Study
CRE	Creatinine
CTEPP	Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants Study
CTEPP-NC	CTEPP Study, North Carolina homes and daycares
CTEPP-NC d	CTEPP Study, North Carolina daycares only
CTEPP-NC DAYCARE	CTEPP Study, North Carolina daycares only
CTEPP-NC h	CTEPP Study, North Carolina homes only
CTEPP-NC HOME	CTEPP Study, North Carolina homes only
CTEPP-OH	CTEPP Study, Ohio homes and daycares
CTEPP-OH d	CTEPP Study, Ohio daycares only
CTEPP-OH DAYCARE	CTEPP Study, Ohio daycares only
CTEPP-OH h	CTEPP Study, Ohio homes only
CTEPP-OH HOME	CTEPP Study, Ohio homes only
DAP	Dialkylphosphate

Daycare / DAYCARE	Feasibility of Macroactivity Approach to Assess Dermal Exposure Study
Dep Coup	Deposition coupon
DC	Deposition coupon
DCHD	Duval County Health Department
DEET	N,N-diethyl-meta-toluamide
DIYC	Dietary Intake of Young Children Study
EOSHI	Environmental and Occupational Health Sciences Institute Study
Food	Food Transfer Studies
FQPA	Food Quality Protection Act
GC/ECD	Gas Chromatography/Electron Capture Detector
GC/MS	Gas Chromatography/Mass Spectroscopy
GLM	Generalized linear model
GM	Geometric mean
GSD	Standard deviation of the geometric mean
HUD	US Department of Housing and Urban Development
HVS3	High Volume Small Surface Sampler
ICC	Intraclass Correlation Coefficient
IMP / IMPy	2-Isopropyl-6-methyl-4-pyrimidinol
IPA	Isopropyl alcohol
IPA Wipe	Isopropyl alcohol wipe
JAX	Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposures in Children Living in Jacksonville, Florida Study
JAX-AG	JAX Study, Aggregate Exposure Assessment phase
JAX-AGG	JAX Study, Aggregate Exposure Assessment phase
JAXAGGREGATE	JAX Study, Aggregate Exposure Assessment phase
JAX-SC	JAX Study, Screening phase
JAX-SCR	JAX Study, Screening phase
JAXSCREENING	JAX Study, Screening phase
LOD	Limit of detection
LWW	Lioy-Weisel-Wainman wipe sampler
Max	Maximum
MCPA	(4-chloro-2-methylphenoxy)acetic acid
MDA	Malathion dicarboxylic acid
MDL	Minimum detection limit
MGK 264	N-octyl bicycloheptene dicarboximide
Min	Minimum
MNCPES / MN	Minnesota Children's Pesticide Exposure Study
MPA	2-methyl-3-phenylbenzoic acid
N	Sample size
NC Daycare	CTEPP Study, North Carolina daycares only
NC DC	CTEPP Study, North Carolina daycares only
NC HM	CTEPP Study, North Carolina homes only
NC Home	CTEPP Study, North Carolina homes only

NC	North Carolina
NERL	National Exposure Research Laboratory
NHANES	National Health and Nutrition Examination Survey Study
NHEXAS-AZ	National Human Exposure Assessment Survey in Arizona
NOPES	Non-Occupational Pesticide Exposure Study
NRMRL	National Risk Management Research Laboratory
OCHP	Office of Children's Health Protection
OH Daycare	CTEPP Study, Ohio daycares only
OH DC	CTEPP Study, Ohio daycares only
OH HM	CTEPP Study, Ohio homes only
OH Home	CTEPP Study, Ohio homes only
OH	CTEPP Study, Ohio
OP	Organophosphate
OPP	Office of Pesticide Programs
OPPT	Office of Pollution Prevention and Toxics
ORD	Office of Research and Development
P25	25th percentile
P50	Median / 50th percentile
P75	75th percentile
P95	95th percentile
PBPK	Physiologically-Based Pharmacokinetic Model
PET	A Pilot Study Examining Translocation Pathways Following a Granular Application of Diazinon to Residential Lawns Study
PUF	Polyurethane foam
PYR	Pyrethroid
REJV	Residential Exposure Joint Venture
RTI	Research Triangle Institute
SD	Standard deviation of the arithmetic mean
SHEDS	Stochastic Human Exposure and Dose Simulation Model
STAR	Science to Achieve Results
TCPY / TCP / TCPy	3,5,6-Trichloro-2-pyridinol
TE	Transfer Efficiency
TEST / TESTHOUSE / Test House	The Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the US EPA Indoor Air Quality (IAQ) Research House Study
TESTHOUSE Pre	Test House Study, pre-application day only
t-Permethrin	*trans*-Permethrin
t-Perm	*trans*-Permethrin
trans-P	*trans*-Permethrin
Transfer	Characterizing Pesticide Residue Transfer Efficiencies
US CPSC	US Consumer Product Safety Commission
US EPA	U.S. Environmental Protection Agency
VTD	Virtual Timing Device

1.0 INTRODUCTION

1.1 Background

The U.S. Environmental Protection Agency (U.S. EPA) has pledged to increase its efforts to provide a safe and healthy environment for children by ensuring that all EPA regulations, standards, policies, and risk assessments take into account special childhood vulnerabilities to environmental toxicants. Children are behaviorally and physiologically different from adults. Their interaction with their environment, through activities such as playing on floors, and mouthing of hands and objects, and handling of food, may increase contact with contaminated surfaces. Proportionately higher breathing rates, relative surface area, and food intake requirements may increase exposure. Differences in absorption, metabolism, storage, and excretion may result in higher biologically effective doses to target tissues. Immature organ systems may be more susceptible to toxicological challenges. Windows of vulnerability, when specific toxicants may permanently alter the function of an organ system, are thought to exist at various stages of development.

Children are exposed to a wide variety of chemicals in their homes, schools, daycare centers, and other environments that they occupy. The chemicals to which they are exposed may originate from outdoor sources, such as ambient air contaminants, indoor sources such as building materials and furnishings, and from consumer products used indoors. One category of consumer products to which children may be exposed is pesticides that are used to control roaches, rats, termites, ants, and other vermin. Despite widespread residential and agricultural use of pesticides, only limited measurement data are available for pesticide levels in environments that children occupy and little is known about the factors that impact children's exposures to pesticides. The Food Quality Protection Act (FQPA) of 1996 requires EPA to upgrade the risk assessment procedures for setting pesticide residue tolerances in food by considering the potential susceptibility of infants and children to both aggregate and cumulative exposures to pesticides. Aggregate exposures include exposures from all sources, routes, and pathways for individual pesticides. Cumulative exposures include aggregate exposures to multiple pesticides with the same mode of action for toxicity. FQPA requires risk assessments to be based on exposure data that are of high quality and high quantity or on exposure models using factors that are based on existing, reliable data.

EPA's Office of Research and Development (ORD) is responsible for conducting research to provide the scientific foundation for risk assessment and risk management at EPA. In 2000, ORD released its *Strategy for Research on Environmental Risks to Children* addressing research needs and priorities associated with children's exposure to environmental pollutants and providing a framework for a core program of research in hazard identification, dose-response evaluation, exposure assessment, and risk management.

The National Exposure Research Laboratory (NERL) in ORD is working to achieve three specific objectives of the *Strategy* through its children's exposure research program: (1) develop improved exposure assessment methods and models for children using existing information; (2) design and conduct research on age-related differences in exposure, effects, and dose-response relationships to facilitate more accurate risk assessments for children; and (3) explore

opportunities for reducing risks to children. After an exhaustive review of the volume and quality of the data upon which default assumptions for exposure factors are based (Cohen Hubal *et al.,* 2000a), a framework for systematically identifying the important sources, routes, and pathways for children's exposure was developed (Cohen Hubal *et al.*, 2000b).

This framework (Figure 1.1), based on a conceptual model for aggregate exposure, provides the foundation for a protocol for measuring aggregate exposures to pesticides (Berry *et al.,* 2001) and for developing sophisticated stochastic models (Zartarian *et al.,* 2000). Using the framework, four priority research areas, representing critical data gaps in our understanding of environmental risks to children, have been identified:

(1) Pesticide use patterns;
(2) Spatial and temporal distribution in residential dwellings;
(3) Dermal absorption and indirect (non-dietary) ingestion (including micro- and macro-activity approaches); and
(4) Direct ingestion.

Several targeted studies were designed and conducted to address these research needs. These include laboratory studies, small pilot field studies, and large collaborative observational studies. These studies aimed to: (1) evaluate methods and protocols for measuring children's exposure, (2) collect data on exposure factors to reduce the uncertainty in exposure estimates and risk assessments, and (3) collect data for use in exposure model development and evaluation.

1.2 Purpose of the Report and Intended Audience

This document is a comprehensive summary report of data collected under or otherwise related to the NERL children's exposure research program. Data are compared across studies and across compounds to identify or evaluate important factors influencing exposures along each relevant pathway. Summary statistics, comparative analyses, and spatial and temporal patterns are presented to address previously identified data gaps. The primary purpose of this document is to identify factors that are most important for children's exposures to pesticides. The objectives of this document are to:

- Compare results across studies in order to identify trends or similar observations that might provide a better understanding of the factors affecting children's exposures;
- Describe recent children's exposure studies conducted or funded by NERL, including descriptions of the parameters measured and the measurement methods;
- Provide concentration data and summary statistics for comparison of the studies; and
- Present highlights of the results of the studies.

The document was completed with input from staff in the EPA Program Offices, NERL researchers, and Science to Achieve Results (STAR) program grantees who gathered at the US EPA National Exposure Research Laboratory's Workshop on the Analysis of Children's Measurement Data (Tulve *et al.,* 2006) in September 2005 to discuss data presented in a draft summary report, assess the suitability of the data for testing key hypotheses, and propose additional analyses.

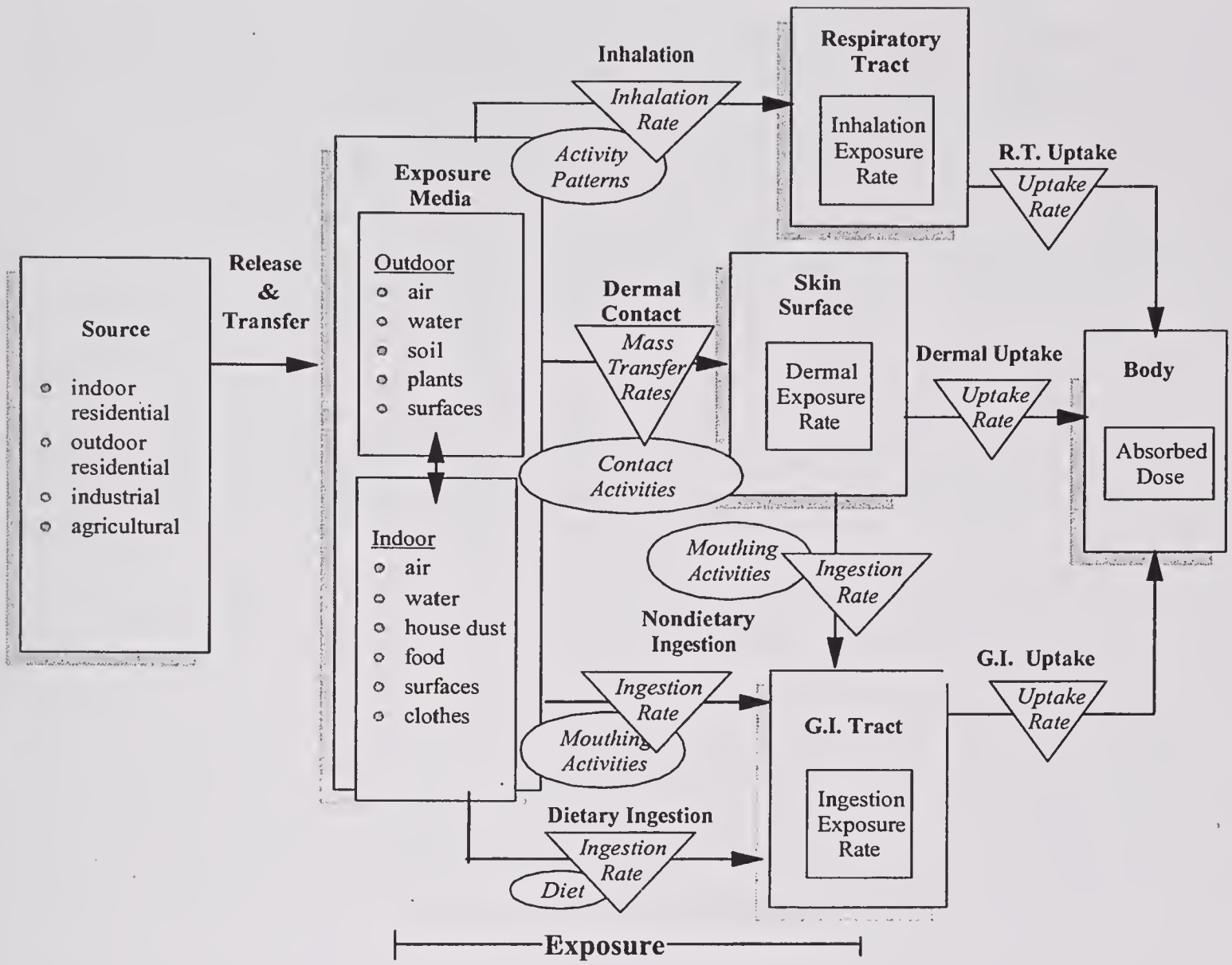


Figure 1.1 Modeling framework for children's pesticide exposure from Cohen Hubal *et al.* (2000b).

The document is intended for an audience of exposure scientists, exposure modelers, and risk assessors. Exposure scientists will find a useful evaluation of available sampling methods for all media relevant to children's exposures. Exposure modelers will be able to use the data to develop or improve probabilistic multimedia, multi-pathway human exposure models. Most significantly, the report may be used by EPA Program offices such as the Office of Pesticide Programs (OPP), the Office of Pollution Prevention and Toxics (OPPT), and the Office of Children's Health Protection (OCHP) to enhance the Agency's risk assessment activities by replacing default assumptions with high-quality, real-world data. Fewer default assumptions will lead to more accurate assessments of exposure and risk and will bolster ensuing risk reducing actions. Furthermore, by examining relationships among application patterns, exposures, and biomarkers for multiple compounds from different classes of pesticides, this report contributes to the development of more reliable approaches for assessing cumulative exposure. Some of the analyses and comparisons that are presented in this summary report include the following:

- Comparison of concentrations
- Spatial variability
- Temporal variability
- Regional comparisons
- Urban versus rural
- Home versus daycare
- Indoor versus outdoor
- Parent compound versus metabolite
- Effect of physical and chemical properties
- Impact of air exchange rate
- Effect of surface type
- Effect of surface concentration
- Effect of sampling method

Comparisons between studies may involve different numbers of measurements, different sampling strategies and methods, and different chemical analysis methods

1.3 Structure of the Report

This document presents data from studies to evaluate children's exposure to pesticides, spanning from pesticide use patterns, through concentrations in exposure media, to biological markers of exposure. The exposure media are listed in an order that roughly mirrors the complexity of the exposure mechanism; that is, beginning with inhalation exposure and ending with dermal exposure. At the beginning of each section, available data from the relevant studies are listed. Results are presented in tables and graphs to illustrate the available data and to facilitate comparisons both across studies and across pesticides.

Throughout the document, lognormal probability plots ("logplots") and box-and-whisker plots ("box plots") are used to graphically depict and compare distributions of concentrations or surface loadings. The logplots are used to compare results only from large observational field studies and the boxplots are used to compare results from the focused studies against each other and against the large observational field studies. In the lognormal probability plot, the ordered

values of the measured concentration are plotted on a log-scale vertical axis, and the percentiles of the theoretical normal distribution are plotted on the horizontal axis. If the points in the plot form a nearly straight line, the data are approximately lognormal. The box-and-whisker plot is actually a group of side-by-side box-and-whisker plots along the x-axis, each representing a different study. The upper whisker extends to the maximum value, the upper edge of the box represents the 75th percentile, the line inside the box represents the median (50th percentile), the lower edge of the box represents the 25th percentile, and the lower whisker extends to the minimum value. Note that the vertical axis is log-scale.

1.4 Data Treatment

Values that are below the method detection limit (MDL) are common in environmental data sets. All values above the MDL are statistically different from zero; however, values near the MDL are generally less accurate than those much higher than the MDL. Laboratories often report a second limit, the Method Quantitation Limit (MQL), as the smallest amount that can be *reliably quantified* in a sample. Despite the higher relative uncertainty in values between the MDL and the MQL, all values above the MDL are retained for the purposes of this document. Values below the MDL are treated using simple substitution, wherein they are replaced with a fraction of the detection limit (MDL/$\sqrt{2}$), a common practice originally proposed by Hornung and Reed (1990). These substituted values are used in all statistical analyses performed specifically for this report and are presented in all data plots, except for lognormal probability plots, in which these substituted values were judged by the authors to be misleading. Detection frequencies (that is, the percent of measurements above the MDL) are presented for each compound by each relevant sampling method at the beginning of each chapter.

Sampling weights are available for all of the large-scale observational field studies, but, unless otherwise noted, only unweighted concentrations are presented in this report. Summary statistics based on unweighted observations may not provide as valid an estimate of true study population values as those based on weighted observations, but are used nonetheless to maintain consistency in comparisons with studies for which weights are not available. In all cases where a statistical test was done to assess differences, the name of the test and the resulting p-value are presented.

1.5 Description of the Studies and Data Collected

Data are included in this report from the following studies. (The acronyms in parentheses are used in the Tables and Figures of this report.)

- National Human Exposure Assessment Survey in Arizona (NHEXAS-AZ)
- Minnesota Children's Pesticide Exposure Study ("MNCPES")
- Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants ("CTEPP")
- First National Environmental Health Survey of Child Care Centers ("CCC")
- Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposures in Children Living in Jacksonville, Florida ("JAX")
- Center for the Health Assessment of Mothers and Children of Salinas Quantitative Exposure Assessment Study ("CHAMACOS")

- Children's Pesticide Post-Application Exposure Study ("CPPAES")
- Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the US EPA Indoor Air Quality Research Test House ("Test House")
- Pilot Study Examining Translocation Pathways Following a Granular Application of Diazinon to Residential Lawns ("PET")
- Dietary Intake of Young Children ("DIYC")
- Characterizing Pesticide Residue Transfer Efficiencies ("Transfer")
- Food Transfer Studies ("Food")
- Feasibility of Macroactivity Approach to Assess Dermal Exposure ("Daycare")

All studies involving children were observational research studies, as defined in 40 CFR Part 26.402. All study protocols and procedures to obtain the assent of the children and informed consent of their parents or guardians were reviewed and approved by independent Institutional Review Boards (IRBs) and complied with all applicable requirements of the Common Rule (45 CFR 46) regarding additional protections for children (Subpart D). Further, all protocols regarding recruitment and treatment of participants were reviewed by the EPA Human Subjects Research Review Official (HSRRO) to assure compliance with the Federal Policy for the Protection of Human Subjects.

The studies discussed in the report included large observational studies, such as NHEXAS-AZ, MNCPES, CTEPP, and CCC, small pilot-scale observational studies (*e.g.*, JAX, CPPAES, DIYC, CHAMACOS, and Daycare), and laboratory studies (*e.g.*, Test House, Transfer, and Food).

- MNCPES, NHEXAS-AZ, CTEPP, and CCC were large observational exposure measurement studies with survey designs that involved random sampling. The CCC study was a nationwide survey and the others had a regional focus. Sampling weights are available for all of these studies, but, unless noted otherwise, only unweighted concentrations are presented in this report.
- The small pilot-scale observational studies are small-scale field studies, such as JAX and CHAMACOS, which were performed to evaluate methods for conducting aggregate exposure assessments for pesticides and to collect preliminary data that could be used to assist in the design of larger observational studies. Like the large observational studies, some of these smaller studies included measurements of multiple chemicals in multiple media.
- The laboratory studies consisted of experiments under controlled conditions to evaluate factors affecting transfer from surfaces (Transfer and Food studies). The Test House study investigated the fate and transport of chlorpyrifos following a crack and crevice application and provided valuable information on spatial and temporal variability of surface concentrations in the absence of human activity.

During these studies, the following types of measurements were collected (not all types of samples were collected in all studies):

- Air (indoor and outdoor)
- Soil
- House dust – Floors (carpet and hard surface)
- Surface wipes (including eating and food preparation surfaces)
- Transferable residues (*e.g.*, polyurethane foam roller, C18 press)
- Hand wipes
- Dermal surrogates (cotton garment and socks)
- Duplicate diet (solid food, beverages)
- Handled food
- Urine

Information was also typically collected by questionnaire on:

- Housing characteristics
- Participant characteristics
- Children's activities (timelines and logs)
- Recent pesticide use

The types of media sampled and questionnaires administered in each study are listed in Table 1.1. Other than the pesticide inventory and use questionnaires, questionnaire data are not the focus of this document.

1.6 Pesticides of Interest to this Report

The studies presented here were performed when a number of organophosphate and pyrethroid pesticides were in use; thus numerous pesticides from various chemical classes (including insecticides and herbicides) were measured. All measured insecticides (and insecticides synergists) are listed in Table 1.2, although not all of the studies collected data for all of the insecticides listed. To reduce complexity, this report focuses on the most commonly detected organophosphate and pyrethroid insecticides:

- Chlorpyrifos
- Diazinon
- Permethrin
- Cyfluthrin

Table 1.1 Available media, participant characteristics, and activities by study.

	NHEXAS-AZ	MNCPES	CTEPP	CCC	JAX SCREENING	JAX AGGREGATE	CHAMACOS	CPPAES	TESTHOUSE	PET	DIYC	DAYCARE
Air - Indoor	✓	✓	✓			✓	✓	✓	✓	✓	✓	
Air - Outdoor	✓	✓	✓			✓	✓				✓	
House Dust	✓		✓				✓					
Surface Residue Wipes	✓		✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
LWW Surface Sampler		✓						✓				
Transferable Residues	✓	✓	✓	✓		✓	✓		✓	✓	✓	✓
Hand Wipes	✓	✓	✓					✓		✓	✓	✓
Cotton Garments/Socks						✓	✓	✓		✓		✓
Soil		✓	✓	✓			✓			✓		
Duplicate Diet	✓	✓	✓			✓					✓	
Handled Foods											✓	
Urine	✓	✓	✓		✓	✓		✓		✓	✓	
Housing Characteristics	✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	
Participant Characteristics	✓	✓	✓	✓	✓	✓		✓		✓	✓	✓
Children's Activities	✓	✓	✓			✓		✓		✓	✓	✓
Recent Pesticide Use		✓	✓	✓	✓	✓						
Pesticide Inventory		✓			✓	✓						

Table 1.2 Pesticides and metabolites measured in the studies.

Pyrethroid	Organophosphorus		Other
Allethrin	Acephate	Ethyl parathion	Fipronil
Bifenthrin	Azinphos-methyl	Fonofos	Piperonyl butoxide
Cyfluthrin [a]	Chlorpyrifos [a]	Malathion	TCPy [a b]
Cyhalothrin	Chlorpyrifos-oxon	Malathion-oxon	IMP [a c]
Cypermethrin	Demeton-S	Methamidophos	3-PBA [a d]
Deltamethrin	Diazinon [a]	Methidathion	
Esfenvalerate	Diazinon-oxon	Methyl-parathion	
Permethrin [a]	Dichlorvos	Mevinphos	
Pyrethrins	Dimethoate	Naled	
Resmethrin	Disulfoton	Phosmet	
Sumithrin	Ethion		
Tetramethrin			
Tralomethrin			

[a] Pesticides and metabolites of primary interest in this document
[b] 3,5,6-Trichloro-2-pyridinol, a selective metabolite of chlorpyrifos
[c] 2-Isopropyl-4-methyl-6-hydroxypyrimidine, a specific metabolite of diazinon
[d] 3-phenoxybenzoic acid, a metabolite common to many pyrethroids

1.7 Summary Descriptions of the Studies

Individual study details are listed in Appendix B. Journal articles presenting results of these studies are listed in the Bibliography. The studies are summarized below.

The National Human Exposure Assessment Survey in Arizona (NHEXAS-AZ) was performed in collaboration with the University of Arizona, the Illinois Institute of Technology, and Battelle Memorial Institute. Probability-based samples were collected in each of Arizona's 15 counties from December 1995 to March 1997. Although 176 households participated, this report only includes data from 21 households with children ages 6-12 as primary participants. Environmental samples included indoor and outdoor air (3-day integrated samples), personal air (1-day), vacuumed surface dust, and window sill wipes. Personal samples included 24-hour duplicate diet and hand wipes. Biological samples consisted of urine samples (first morning void). Baseline and follow-up questionnaires and time-activity diaries captured activity patterns. Two pesticides (and their metabolites) were of primary interest, namely chlorpyrifos (TCPy) and diazinon, and two pesticides (and their metabolites) were of secondary interest, namely malathion (MDA) and carbaryl (1-naphthol).

The Minnesota Children's Pesticide Exposure Study (MNCPES) was an observational measurement study performed in collaboration with Research Triangle Institute (RTI), the Environmental and Occupational Health Sciences Institute (EOHSI), the Minnesota Department of Health, and the University of Minnesota. A telephone survey and in-home interviews were used to collect data on pesticide storage and use patterns from 308 households in both urban centers (Minneapolis/St. Paul) and rural counties (Goodhue and Rice) during the summer of 1997. Probability-based sampling weights were developed and intensive environmental and personal monitoring were performed for 102 children, ages 3-13. Households reporting more frequent pesticide use were oversampled. Environmental samples included personal, indoor, and outdoor air (6-day integrated), surface dust (wipe and press), surface soil, and tap water. Personal samples included solid food (4-day composite), beverages (4-day composite), hand rinse, and first morning void urine (days 3, 5, and 7). In addition to questionnaires and diaries, videotaping was performed in a subset of 20 homes. Four primary pesticides (and their metabolites), namely chlorpyrifos (TCPy), atrazine (atrazine mercapturate), malathion (malathion dicarboxylic acid), and diazinon, and 14 secondary pesticides were measured, along with 13 polycyclic aromatic hydrocarbons (PAHs).

The Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) Study (Morgan *et al.,* 2004) was performed in collaboration with Battelle Memorial Institute as an observational study of preschool children's exposure to contaminants in their everyday environments (*i.e.*, homes and daycare centers). Monitoring was performed from July 2000 to March 2001 in North Carolina (spanning summer, fall, and winter) and from April 2001 to November 2001 in Ohio (spanning spring, summer, and fall). The study population consisted of 257 children, ages 18 months to five years, and their primary adult caregivers (130 children, 130 homes, and 13 daycare centers in North Carolina; 127 children, 127 homes, and 16 daycare centers in Ohio). Samples were collected over a 48-hr period at each home and daycare center, including indoor air, outdoor air, floor dust, soil, hand wipe, solid food, liquid food, and urine. Supplemental information included a recruitment survey, a house/building characteristics survey, pre- and post monitoring questionnaires, and activity and food diaries. In addition, 20% of the

OH participants were videotaped at home for about 2 hours. Additional samples (hard floor and food preparation surface wipes and transferable residues) were collected if the participant reported indoor or outdoor applications of pesticides within 7 days of the monitoring period.

The First National Environmental Health Survey of Child Care Centers (CCC) was performed in collaboration with HUD (US Department of Housing and Urban Development) and CPSC (US Consumer Product Safety Commission). Samples were collected from August through October (summer and fall) 2001, at 168 randomly-selected child care centers nationwide. Many facilities reported recent pesticide application (either by professionals or by employees). Samples included soil, surface wipes, and transferable residues (C18 Press). A multi-residue chemical analysis method was used to measure a large suite of current-use pesticides. The study aimed to collect data on pesticide use practices and to characterize the distributions of pesticide concentrations in a nationally-representative sample of child care centers in the U.S.

The study titled Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposures in Children Living in Jacksonville, Florida (JAX) was performed in collaboration with CDC (Centers for Disease Control and Prevention) and DCHD (Duval County Health Department) in Jacksonville (Duval County), Florida, from August through October (summer and fall) 2001. The CDC performed a biomonitoring study to measure metabolites of organophosphate and pyrethroid pesticides in a sample of 200 children who were 4-6 years of age. The DCHD conducted a home screening survey in a subset of 42 of the homes. The screening phase employed a pesticide screening inventory, surface wipes, and urine collection. The EPA conducted an observational study in a subset of nine of the homes to evaluate sampling and analysis methods and protocols for conducting aggregate exposure estimates for children. The aggregate exposure study included the pesticide screening inventory, surface wipes, indoor and outdoor air, cotton garment, duplicate diet, and transferable residue measurements, a time activity diary, and a urine sample.

The Center for the Health Assessment of Mothers and Children of Salinas (CHAMACOS) Quantitative Exposure Assessment Study was a collaboration with the University of California at Berkeley. This observational study was performed in homes of agricultural workers living in Salinas, California. Twenty households with children ages 5 months to 3 years old (10 female and 10 male) were monitored during the period of June to October (summer and fall) 2002. Samples were collected over a 24-hour monitoring period and included indoor and outdoor air, house dust, transferable residues from floors (surface wipes and press samples), transferable residues from toys (surface wipes), urine, and cotton union suits and socks. A time/activity diary was also administered. The objective of the study was to evaluate sampling and analysis methods and study protocols that might be applied in larger studies such as the National Children's Study.

The Children's Pesticide Post-Application Exposure Study (CPPAES) was a collaborative field study with EOHSI (Environmental and Occupational Health Sciences Institute) in urban New Jersey over a two-year period stretching from April 1999 to March 2001. Ten homes with children 2-5 years of age participated. Each of the homes had a professional "crack and crevice"-type application of a chlorpyrifos-based formulation at the time of the study, but only trace amounts of chlorpyrifos were applied in three of the homes. The monitoring period typically lasted for two weeks with pre- and multiple post-application samples. Sampling was

comprehensive with indoor air, deposition coupons, surface samples (LWW, Lioy-Weisel-Wainman sampler), toys, hand wipes, urine, air exchange rate, and time activity diary data collected throughout the study, and additional samples consisting of surface wipes, dermal wipes, cotton garments, and videotaped activities collected on the second day of the study.

A field laboratory study titled the Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the US EPA Indoor Air Quality Research Test House (Test House) was performed in collaboration with the National Risk Management Research Laboratory (NRMRL). The Test House is an unoccupied three-bedroom house in Cary, NC. The study investigated the translocation of chlorpyrifos and the spatial and temporal variability of chlorpyrifos levels in air and on surfaces following a professional "crack and crevice"-type application onto the floor and cabinetry of a kitchen. Samples included air, polyurethane foam (PUF) roller, carpet sections, C18 surface press, and surface wipes from multiple rooms. Samples were collected pre-application and on days 1, 3, 7, 14 and 21 post-application.

The Pilot Study Examining Translocation Pathways Following a Granular Application of Diazinon to Residential Lawns (PET) was performed during spring 2001 in six residential homes within a 50-mile radius of Durham, NC. Measurements were performed at homes where a homeowner applied a turf application of a granular formulation of diazinon. Sampling included indoor air (multiple rooms), PUF roller (outdoor and indoor), soil, doormat, high-volume small surface sampler (HVS3), dermal surrogate (cotton gloves), urine (adult and child), dog fur clippings, dog paw wipes, dog blood, and videotaping (15-min). Samples were collected pre-application and 1, 2, 4 and 8 days post-application. A feasibility study was also performed in a single home. The study focused on pesticide translocation and exposure pathways.

The Dietary Intake of Young Children (DIYC) study was a small observational field study in collaboration with RTI. It included three homes where diazinon had been applied (two homes with commercial crack and crevice applications and one home with non-professional application) and took place between November 1999 and January 2000 (fall and winter). Collected samples included indoor air, outdoor air, surface wipes, hand wipes, surface press, food press, food samples, PUF roller, entry wipe, and urine. A primary goal of the study was to evaluate the potential for exposure to pesticides due to food preparation and handling in the home.

The Feasibility of the Macroactivity Approach to Assess Dermal Exposure (Daycare) study was another collaboration with RTI (Cohen Hubal *et al.*, 2006). In this field study, nine daycare centers were identified that reported routine pesticide applications as part of the center's pest control program. In each daycare, screening sampling was conducted to evaluate the distribution of transferable pesticide residues on floor surfaces in the area where children spent the most time. One daycare was selected for more intensive monitoring during the summer of 2001, following a series of regularly scheduled (monthly) applications. Surface sampling and videotaping of activities were conducted simultaneously with dermal surrogate (cotton garment) sampling to calculate dermal transfer coefficients.

The Characterizing Pesticide Residue Transfer Efficiencies (Transfer) studies evaluated parameters that are believed to affect residue transfer from surface-to-skin, skin-to-object, skin-to-mouth, and object-to-mouth. The collaboration with Battelle was a series of controlled laboratory studies using fluorescent tracers as surrogates for pesticide residues. The protocol

involved applying fluorescent tracers to surfaces of interest as a residue at levels typical of residential pesticide applications, and then conducting controlled transfer experiments varying six parameters in a systematic fashion. Repetitive contacts with contaminated surfaces were used to measure the following transfers: hand to clean surface, hand to washing solution, and hand to mouth. In the mouthing trials, mouthing was simulated using saliva-moistened PUF material to measure mass of tracer transferred. Laboratory evaluations were performed to relate transfer of tracer to transfer of pesticides (Ivancic *et al.*, 2004; Cohen Hubal *et al.*, 2005).

The Food Transfer Studies were controlled laboratory experiments investigating pesticide transfer from household surfaces to foods and evaluating factors that have been identified as important, including surface type, duration of contact, surface loading, and contact pressure (applied force). Organophosphate, pyrethroid, and pyrazole insecticides were applied onto various household surfaces using a customized spray chamber. Pesticide transfer efficiencies were measured for three different foods, with standardized surface contact areas. Amounts of pesticide residue transferred to foods were compared to the amounts removed using surface wipes. Transfer efficiency (TE) was defined as the amount of pesticide recovered from the food item divided by the pesticide concentration or loading level.

1.8 Exposure and Dose Models

It is neither within the scope nor the intention of this report to provide a detailed discussion of the exposure and dose models that have been developed using these data or applied to these data. However, since human exposure research progresses through an iterative series of models and measurements, it is often necessary to refer to these models. Models are constructed using current knowledge and are subsequently used to identify areas of greatest uncertainty. Modeled results are used to direct the focus of the measurement studies to address those identified uncertainties. As newly collected data yields new knowledge, models are refined and the entire process repeats. At each iteration, real-world data replace default assumptions to produce more accurate assessments of exposure and risk. Throughout this document models are mentioned. "Algorithms" are the set of deterministic mathematical expressions developed in the *Draft Protocol for Measuring Children's Non-Occupational Exposure to Pesticides by all Relevant Pathways* (Berry *et al.*, 2001) to assess exposure by each route as a function of concentration and various exposure factors. The Stochastic Human Exposure and Dose Simulation (SHEDS) model (Zartarian *et al.*, 2000) is a physically-based, probabilistic model that predicts multimedia/ multipathway exposures and doses incurred eating contaminated foods, inhaling contaminated air, touching contaminated surfaces, and ingesting residues from hand- or object-to-mouth activities. It combines information on pesticide usage, human activities, environmental concentrations, and exposure and dose factors using Monte Carlo methods. The Exposure Related Dose Estimating Model (ERDEM) (Blancato *et al.*, 2004) is a physiologically-based pharmacokinetic (PBPK) model used to make reliable estimates of the chemical dose to organs of animals or humans. It solves a system of differential equations that describes the organ system, directly addressing the uncertainties of making route-to-route, low-to-high exposure, and species-to-species extrapolations when there are exposures to one or to multiple chemicals. The Children's Dietary Intake Model (CDIM) (Hu *et al.*, 2004) estimates total dietary exposure of children to chemical contaminants by accounting for excess dietary exposures caused by chemical contaminant transfer from surfaces and/or hands to foods prior to consumption.

2.0 PESTICIDE USE PATTERNS

Very limited data are available to EPA researchers on what pesticides are currently being used in non-occupational environments, where they are being used, and the frequency of use. The EPA has not conducted a large scale survey to collect data on pesticide use patterns in the U.S. since 1990, but use patterns are believed to have substantially changed since that time. The children's observational studies described in this report collected information on household pesticide use as ancillary information that could be used to address this serious data gap. Despite the limited coverage of geographic regions, a relatively small number of study participants, and the general lack of knowledge about the active ingredients in brand name products on the part of consumers, valuable information was obtained. The NERL studies described in this section covered a period from 1997 to 2001. The indoor residential use of chlorpyrifos was cancelled while data collection was still ongoing in several studies (JAX, CCC, and CTEPP).

The pesticides available to consumers or professionals for use in residential settings have changed over time. By the late 1980s the use of most organochlorine pesticides (*e.g.*, DDT, chlordane, dieldrin, and heptachlor) was severely restricted in the U.S. The organophosphate (OP) insecticides (*e.g.*, malathion, chlorpyrifos, and diazinon), appealing for their high insect toxicity, low costs, and low likelihood of pest resistance, quickly filled the void and became the pesticides of choice for both consumers and professional pest control operators (Karalliedde *et al.*, 2001). The popularity of pyrethroid insecticides increased throughout the 1990s because of the following favorable properties: higher insecticidal toxicity, lower mammalian toxicity, and more rapid environmental degradation (Baker *et al.*, 2004). Passage of the Food Quality Protection Act of 1996 led the EPA to consider aggregate childhood pesticide exposure. The OPs were the first class of pesticides whose tolerances were reassessed, leading to withdrawal of the registrations for indoor applications of chlorpyrifos and diazinon in 2001 and 2002, respectively, because of concern regarding the risk to children. Consequently, pyrethroids have become the leading residential insecticides. While household use of diazinon and chlorpyrifos is now restricted, these and other OPs are still widely used in agriculture, and some structural uses for chlorpyrifos, including the treatment of house foundations, are still approved.

2.1 Sources of Information

Important sources of information on pesticide use patterns in non-occupational environments include Market Estimates from EPA's Office of Pesticide Programs (US EPA, 2004), national pesticide usage surveys, the Residential Exposure Joint Venture (REJV), the National Health and Nutrition Examination Survey (NHANES), and published scientific literature.

The Office of Pesticide Programs uses proprietary data sources in producing "Market Estimates" of pesticide sales and use in various market sectors. According to their estimates, the annual amount of insecticide active ingredients used in the home and garden sector declined from 24 million pounds in 1982, to less than 13 million pounds in 1988. Although the figure rose to 17 million pounds between 1998 and 2001, it still represents a significant decline from the early 1980s. In contrast, the amount of herbicides applied steadily increased over the same period, nearly doubling from 37 million pounds in 1982 to 71 million pounds in 2001 (US EPA, 2004)

as lawn coverage increased. In 2001, insecticides comprised nearly 60% and herbicides nearly 30% of the home and garden sector expenditures (US EPA, 2004).

The REJV is a program administered by eight pesticide registrants and is designed to provide home pesticide usage information critical for risk assessments on individual active ingredients as well as aggregate and cumulative risk assessments. Pesticide use by over 100,000 households in nine regions of the U.S. is recorded, with a year-long monthly diary of all residential pesticide applications in more than 4000 households. EPA expects to use the results of this comprehensive pesticide use survey to refine or replace many of its residential exposure default assumptions. Access to REJV results is restricted as confidential business information, thus only very limited data are publicly available.

Results from two other national surveys are available: the National Household Pesticide Usage Study (US EPA, 1980; Savage *et al.*, 1981) and the National Home and Garden Pesticide Use Survey (US EPA 1992). The National Household Pesticide Usage Study (1976-1977) found that 91% of the more than 8200 households surveyed reported using pesticides in their home, garden, or yard. According to the slightly more recent National Home and Garden Pesticide Use Survey (1990), 75% of American households reported using insecticides. These surveys, it should be noted, are old and the results are not considered relevant to current pesticide use patterns.

NHANES is an ongoing assessment of the exposure of the U.S. population to environmental chemicals. Beginning with the 1999-2000 cycle, the interview included, at the request of EPA, questions on pesticide applications performed in the past month. According to the most recent survey (2001-2002), 18% of households used insecticides inside the home within the past month, nearly 40% of which were professional treatments. Of households with private yards, 20% reported pesticide applications in the yard during the month, roughly 36% of which were professional treatments. NHANES does not report results by region or by season.

Studies in the open literature can also help to identify pesticide use patterns. Davis *et al.* (1992), Bass *et al.* (2001), Curwin *et al.* (2002), Freeman *et al.* (2004), and Carlton *et al.* (2004) address pesticide use patterns in various geographic locations within the U.S., including Missouri, Arizona, Iowa, Texas, and New York.

A study conducted in Missouri from June 1989 to March 1990 using telephone interviews (Davis *et al.*, 1992) examined pesticide use in the home, garden, and yard. Nearly all 238 families (98%) used pesticides at least one time per year, and two-thirds used pesticides more than five times per year. Pesticides were most commonly used inside the home (80%), followed by in the yard (57%). Flea collars were the most popular pest control product (50%). Diazinon and carbaryl were identified as the two most commonly used active ingredients at that time.

The community-based survey conducted by Bass *et al.* (2001) in Douglas, Arizona in 1999 identified pesticides used in the home, use and storage locations, and disposal methods. All (100%) of the 107 randomly chosen study participants reported using pesticides in the six months prior to the survey, although only 75% reported pest problems. Over 30% used a professional exterminator. A total of 148 pesticide products, representing more than 50 unique active ingredients, were catalogued (1.4 products per home). The synergist piperonyl butoxide

(34%) was most common, followed by pyrethrins (24%), permethrin (18%), allethrin (17%), diazinon (16%), and boric acid (13%). The majority of the pesticides were stored inside the house (70%), typically in the kitchen (45%).

Curwin *et al.* (2002) investigated the differences in pesticide use for 25 farm homes and 25 non-farm homes in Iowa. The target pesticides included atrazine, metolachlor, acetochlor, alachlor, 2,4-D, glyphosate, and chlorpyrifos. Among the non-farm households, 84% used pesticides in their homes or on their lawns or gardens. Only 17% of reported residential pesticide use was by commercial application.

Freeman *et al.* (2004) examined pesticide use patterns during the summer 2000 and winter 2000-2001 seasons among families with very young children in a Texas border community. Pesticide use inside the home showed seasonal variation (82% of homes treated in summer versus 63% in winter). The primary room treated was the kitchen, and the primary structures treated were the floors, lower walls, and dish cupboards. The pesticides used were typically pyrethroid formulations. For nearly all of the pesticides analyzed, no differences were found in pesticide levels in house dust based on family reports of pesticide use in the home or yard.

Carlton *et al.* (2004) surveyed stores in New York City, NY in mid-2003 to determine whether the phase-out of chlorpyrifos and diazinon had been effective and what alternative pesticides were available. The authors found the phase-out to be more effective for chlorpyrifos than for diazinon. The summer after chlorpyrifos sales were to have ended, chlorpyrifos-containing products were found in only 4% of stores that sold pesticides; however, after diazinon sales were to have ended, 18% of stores surveyed, including 80% of supermarkets, still stocked diazinon-containing products. Lower toxicity pesticides, including gels, bait stations, and boric acid, were available in only 69% of the stores and were typically more expensive.

The children's exposure research program collected pesticide use information from homes and daycare centers in the MNCPES, JAX, CTEPP, CCC, and Daycare studies. Information on collection methods is available in Table 2.1. In the context of this report, pesticide use patterns include application frequency, locations, types, methods and active ingredients, as well as pesticides identified in inventories and detected in screenings. The following are highlights of the data collected on pesticide use patterns in these studies. A thorough discussion of MNCPES storage and use patterns is found in Adgate *et al.* (2000).

Table 2.1 Pesticides use information collection methods.

Study	Year	Setting	Inventory	Questionnaire	Screening Wipes
MNCPES	1997	Residence	Brand name, type, EPA registration number, use in past year.	Baseline usage (past year) by participant recollection. Recent use (past week and during monitoring period).	No
CTEPP	2000-2001	Residence and Daycare Center	None	Baseline usage (ever) of insecticides, herbicides, fungicides, or shampoos. Recent use (past week) of any pesticide.	No
CCC	2001	Daycare Center	None	Usage frequency (categories) and locations for specific active ingredients. Questionnaire administered to Center Director or professional applicator.	Yes
JAX	2001	Residence	Brand name, type, EPA registration number. Use in past 6 months, use frequency, use location, and targeted pest noted for each product.	Usage frequency (categories), locations, application methods, and anticipated future use.	Yes
Daycare	2000	Daycare Center	None	Specific active ingredient verified by professional applicator.	Yes

2.2 Application Frequency

The frequency of pesticide application, typically over the past month or year, is generally gathered through questionnaires. Although there is little supporting empirical evidence, it is believed that the frequency of application, along with the form and chemical properties of the pesticide, is an important determinant of indoor air and surface concentrations. It is assumed that residue levels within a residence will rise with increasing pesticide application frequency. Conversely, infrequent pesticide application is assumed to decrease the likelihood of measuring pesticide residues. Arguably, the more frequently pesticide applications occur, the more likely the occupant is to have contact with pesticide residue.

- As presented in Table 2.2, about 20% of study participants in Jacksonville, FL (JAX) reported using pesticides in the past seven days (August to October 2001) compared to 14% in CTEPP-NC (July 2000 to March 2001), 13% in CTEPP-OH (April to November 2001), and only 10% in Minnesota (MNCPES) (May to August 1997). This provides some evidence of a pattern of higher application frequencies in warmer climates. The North Carolina study was the only one to include winter months; the percentage would likely be higher if winter months were excluded.
- About the same proportion (unweighted) of participants that used pesticides in the past month (or planned to use them in the next month) in JAX (51%), used them in the past six months in MNCPES (52%). The percentage of JAX participants is substantially higher than 18-23% reporting insecticide use in the past month in NHANES (Table 2.2).
- Differences according to geographical region become more evident in the CTEPP studies (Table 2.3) when focusing on insecticides and rodenticides, as 74% of the participants in warmer climate North Carolina reported using insecticides or rodenticides compared to only 51% in colder climate Ohio.
- In Minnesota (MNCPES), 88% of the participants used pesticides in the past year, slightly more than the 84% reported by Curwin *et al.* (2002) in Iowa but less than the 98% reported by Davis *et al.* (1992) in Missouri and the 100% reported by Bass *et al.* (2001) in Arizona.
- In the CCC study, 74% of the facilities reported application of pesticides in the last year (63% reported interior and 42% reported exterior applications), and 7% were unsure if any application occurred. Up to 107 pesticide applications per year were reported.
- About a third of the interior and a quarter of the exterior applications in the nationwide CCC study were performed on a monthly basis. In the Daycare study, monthly or more frequent pesticide applications were anecdotally found to be standard practice in the Raleigh-Durham area of North Carolina.

Table 2.2 Proportion (unweighted) of participants reporting pesticide use by study. NHANES participant responses are included for comparison.

Study	Use within the past seven days	Use within the past one month	Use within the past six months
CTEPP-NC	14%	-- [a]	--
CTEPP-OH	13%	--	--
JAX	20%	51%	--
MNCPES	10% [b]	--	52%
NHANES 99-00	--	23% [c]	--
NHANES 01-02	--	18% [c]	--

[a] Information not available
[b] Recruited households
[c] Restricted to use inside of home

Table 2.3 The proportion of CTEPP participants reporting use of four types of pesticides.

Type of Pesticide	North Carolina	Ohio
Herbicides	38%	50%
Insecticides / Rodenticides	74%	51%
Fungicides	6%	4%
Shampoos / Lotions	8%	9%

2.3 Application Locations

Although applied pesticides are redistributed throughout a home following an application, a concentration gradient exists with higher concentrations in the application room and lower concentrations in more distant rooms (Stout and Mason, 2003). Since residential applications may be performed by someone other than the occupant (*e.g.*, professional pest control service, gardener, lawn service, or property management), the occupant may not know which locations were treated.

- In JAX, 58% reported treating all rooms in the home, and 15% reported treating just the kitchen.
- The most commonly treated room in the CCC study was the kitchen (62%), followed by the bathroom (52%) (Figure 2.1). All rooms were treated in 23% of the centers.
- Areas treated by professional crack and crevice applications in CPPAES represented 93% of the homes' living areas.

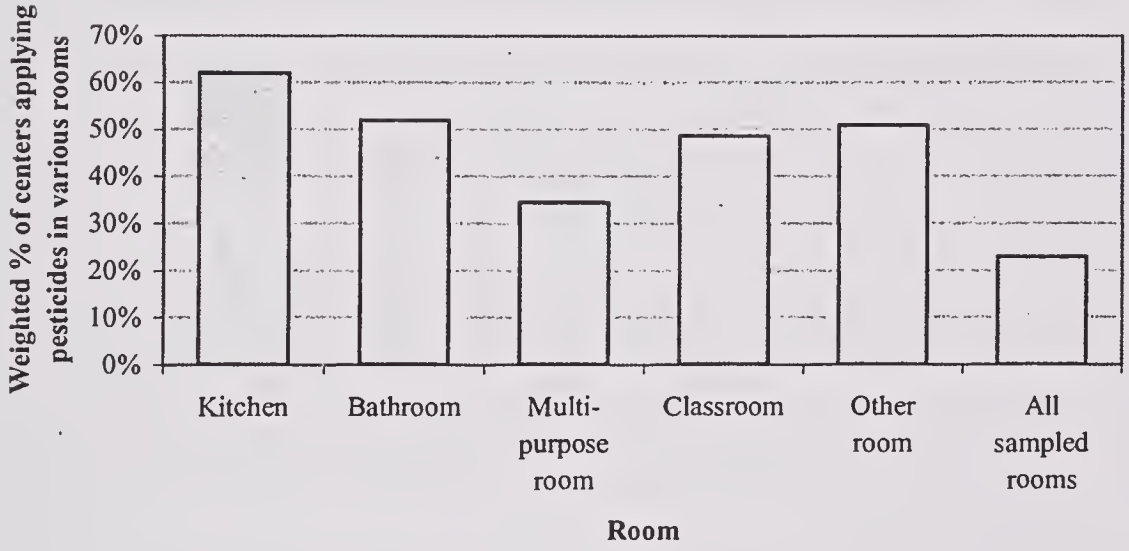


Figure 2.1 Weighted percentage of child care centers reporting treatment of various rooms in the Child Care Centers (CCC) study.

2.4 Application Types and Methods

The three common *types* of pesticide applications in the non-occupational environment are broadcast, total release aerosol, and crack-and-crevice. A broadcast application spreads insecticide onto broad surfaces, typically large sections of walls, floors, ceilings, or in and around trash containers (Rust *et al.*, 1995). Total release aerosols, also known as "foggers" or "bug bombs," contain propellants that release their contents at once to fumigate a large area. Alternatively, a crack-and-crevice application is the application of small amounts of insecticide into areas where pests typically harbor or enter a building. Cracks and crevices are commonly found between cabinets and walls, at expansion joints, and between equipment and floors (Rust *et al.*, 1995). Crack and crevice type applications, which usually produce lower airborne concentrations and surface loadings than broadcast or total release type applications, are favored by professional pest control services.

Method of pesticide application (as differentiated from "type" of application) refers to the equipment or product form used, and may include aerosol sprayer, hand pump sprayer, hose end sprayer, spritz sprayer, hand trigger sprayer, liquid, fogger, gel, granules/dust/powder/pellets, lotion, shampoo, bait station/trap, candle/coil, fly strip, pet collar, and spot-on pet treatment.

- Only very limited information on application type and method was collected in any of the field study questionnaires.
- In CCC, 36% of the interior applications were reported by the center directors as crack and crevice, and only 2% were reported as broadcast. In the Daycare study, all observed pesticide applications were crack and crevice.
- The most common application methods reported in JAX were as follows: 37% hand pump sprayer, 24% aerosol can, 3% fogger, and 3% bait.
- Applications in JAX were more likely to be performed by the respondent or respondent's family member (41%) than by a professional service (35%). These results are similar to NHANES 01-02, where 66% of the survey respondents reported non-professional treatments compared to professional treatments that were reported by 40% of the respondents. These results are also similar to the survey by Bass *et al.* (2001) in Douglas, Arizona, where 30% used professional services.

2.5 Pesticides Identified in Inventories, Records and Wipe Samples

- Pesticide products were found in 86% of the 36 homes inventoried in the JAX study (Table 2.4), with up to three products per household. Pyrethroids were the most common active ingredient (67% of homes), primarily cypermethrin (25%) and allethrin (12%), followed by imiprothrin, pyrethrins, and tralomethrin (all 14%). Only one organophosphate insecticide (diazinon) and one insect repellent (DEET) were found.
- The most commonly inventoried pyrethroids in JAX (Table 2.4) corresponded well with commonly reported pyrethroids in the Residential Exposure Joint Venture (Table 2.5).
- Cataloguing of pesticides in the CCC study (Table 2.6) gave results similar to JAX, with pyrethroid products most commonly identified (second only to products with unknown

active ingredients).

- The finding of 145 application events (Table 2.6) with unidentified active ingredients in the CCC study suggests that tracking of pesticide use in and around daycare facilities may require improved recordkeeping.
- As reported in Adgate *et al.* (2000), pesticide products were found in 97% (weighted) of the MNCPES households. The weighted mean number of pesticide products used per household was 3.1. Participants reported that fewer than 25% of the pesticides inventoried in their homes were used during the past year.
- In MNCPES, DEET-containing products were used in 47% of the homes during the last year (Table 2.7).
- Repellents, pyrethrins and pyrethroids, organophosphates, chlorophenoxy herbicides, and carbamates were present in more than 20% of the MNCPES households (Table 2.7).
- In the Daycare study, professional pest control services applied pyrethroid or pyrethrin pesticides in six of the eight facilities (data not presented). Esfenvalerate was applied in two facilities while cyhalothrin, pyrethrins, cypermethrin, and tralomethrins were each used in one.
- Cypermethrin, *cis*-permethrin, and *trans*-permethrin were detected in over 80% of the surface wipe samples collected in 46 homes in JAX (Table 2.8), consistent with the pesticide inventories. Chlorpyrifos and diazinon, although not identified in the inventories, were present in 89% and 91%, respectively, of the surface wipe samples.
- Permethrin and cypermethrin were the most frequently detected pyrethroid pesticides in both JAX (homes) and CCC (childcare centers) (Table 2.8). Chlorpyrifos and diazinon were the most frequently detected OPs, at frequencies comparable to permethrin.
- As of 2001, the synthetic pyrethroids appeared to be the most frequently used insecticides for indoor applications in homes and child care centers. It is anticipated that their use has become even more common since the cancellation of indoor use registrations of chlorpyrifos (2001) and diazinon (2002).

2.6 Demographic Factors Influencing Applications

- As reported by Adgate *et al.*, (2000), there were no statistically significant differences in the weighted total number of products found or reportedly used in MNCPES based on either population density (urban versus non-urban households) or other socio-demographic factors including race, ethnicity, home type, income, and level of education.
- Chi square analysis of CTEPP data (not presented) found no association between having applied pesticides within the past week and either income class or urban/rural status.

Table 2.4 Pesticides inventoried in 36 households in Jacksonville, FL (JAX) in fall 2001.

Active Ingredient	Pesticide Class	Number of Homes Where Found (% of Homes)
Cypermethrin	Pyrethrins/Pyrethroids	9 (25%)
Allethrin	Pyrethrins/Pyrethroids	8 (22%)
Pyrethrins	Pyrethrins/Pyrethroids	5 (14%)
Imiprothrin	Pyrethrins/Pyrethroids	5 (14%)
Tralomethrin	Pyrethrins/Pyrethroids	5 (14%)
MGK 264 [a]	Synergist	4 (11%)
Permethrin	Pyrethrins/Pyrethroids	4 (11%)
Fipronil	Phenylpyrazole	4 (11%)
Piperonyl butoxide	Synergist	4 (11%)
Hydramethylnon	Aminohydrazone	3 (8%)
Tetramethrin	Pyrethrins/Pyrethroids	3 (8%)
Cyfluthrin	Pyrethrins/Pyrethroids	2 (6%)
Esfenvalerate	Pyrethrins/Pyrethroids	2 (6%)
Prallethrin	Pyrethrins/Pyrethroids	2 (6%)
Bifenthrin	Pyrethrins/Pyrethroids	1 (6%)
DEET	Repellent	1 (6%)
Diazinon	Organophosphate	1 (6%)

[a] N-octyl bicycloheptene dicarboximide

Table 2.5 Most commonly applied pyrethroids in 1217 households with complete 12 month REJV survey data, as reported by Ozkaynak (2005).

Pyrethroid Pesticide	Number of Homes Where Applied (% of Homes)
Permethrin	518 (43%)
Pyrethrins	472 (39%)
Piperonyl Butoxide	461 (38%)
Allethrin	437 (36%)
Tetramethrin	342 (28%)
Phenothrin	293 (24%)
Tralomethrin	279 (23%)
Cypermethrin	163 (13%)
Resmethrin	106 (9%)
Bifenthrin	99 (8%)
Cyfluthrin	46 (4%)
Fenvalerate	37 (3%)
Esfenvalerate	25 (2%)
Deltamethrin	22 (2%)
Prallethrin	13 (1%)
Cyhalothrin	4 (<1%)

Table 2.6 Number of pesticide products applied during one year (2001) in 168 child care centers (CCC), as reported by the center directors and/or professional applicators.

Pesticide Class or Type	Number of Products Applied in Past Year (Unweighted % of All Products)
Unknown	145 (39%)
Pyrethroids	93 (25%)
Phenyl pyrazole or unclassified insecticide	44 (12%)
Pesticide mix	22 (6%)
Fungicide/insecticide	20 (5%)
Organophosphate	10 (3%)
Glueboard/Mouse traps	7 (2%)
Carbamates	6 (2%)
Juvenile hormone mimic insecticide	6 (2%)
Coumarin rodenticides	5 (1%)
Herbicides	3 (1%)
Insecticides	3 (1%)
Unclassified acaricide	3 (1%)
Unclassified insecticide	3 (1%)
Biopesticides	2 (1%)
Pheromone	1 (<1%)
Phosphoramidothioate acaricide	1 (<1%)
Rodenticides	1 (<1%)

Table 2.7 Pesticides inventoried and used in 308 households in Minnesota (MNCPES) in summer 1997 (adapted from Adgate *et al.*, 2000).

Active Ingredient	Pesticide Class	Homes Where Found (Weighted Percent)	Homes Where Used in the Past Year (Weighted Percent)
DEET	Repellent	196 (58%)	162 (47%)
Piperonyl butoxide	Synergist	152 (45%)	91 (25%)
Pyrethrins	Pyrethrins/Pyrethroids	147 (43%)	88 (25%)
MCPA	Chlorphenoxy herbicide	107 (35%)	55 (17%)
Permethrin	Pyrethrins/Pyrethroids	93 (35%)	65 (15%)
Chlorpyrifos	Organophosphate	89 (29%)	55 (17%)
Propoxur	Carbamate	84 (25%)	53 (17%)
MGK 264 [a]	Synergist	83 (25%)	43 (12%)
Allethrin	Pyrethrins/Pyrethroids	81 (24%)	49 (13%)
2,4-D	Chlorphenoxy herbicide	74 (23%)	37 (11%)
Diazinon	Organophosphate	65 (18%)	37 (11%)
Glyphosoate	Aminophosphate	62 (18%)	37 (12%)
Tetramethrin	Pyrethrins/Pyrethroids	62 (18%)	32 (8.5%)
Resmethrin	Pyrethrins/Pyrethroids	60 (20%)	24 (8.1%)
Carbaryl	Carbamate	50 (14%)	24 (5.4%)

[a] N-octyl bicycloheptene dicarboximide

Table 2.8 Detection frequencies of target analytes in soil and wipe samples in the CCC study (weighted) and in screening wipe samples collected in JAX (unweighted).

Compound	CCC			JAX
	% Detect in Soil Samples	% Detect in Floor Wipes	% Detect in Surface Wipes	% Detect in Surface Wipes
PYRETHROIDS				
cis-Allethrin	5	2	0	22
trans-Allethrin	5	2	0	22
Bifenthrin	14	5	4	20
Cyfluthrin	7	7	1	20
lambda-Cyhalothrin	6	7	5	9
Cypermethrin	8	23	9	80
Delta/Tralomethrin	5	2	0	15
Esfenvalerate	9	6	0	30
cis-Permethrin	12	63	48	89
trans-Permethrin	15	64	64	87
Resmethrin	5	3	6	0
Sumithrin	5	2	1	4
Tetramethrin	5	2	0	13
ORGANOPHOSPHATES				
Acephate	50	3	0	7
Azinphos methyl	15	1	0	2
Chlorpyrifos	21	67	76	89
Chlorpyrifos oxon	11	1	1	0
Demeton S	11	0	0	0
Diazinon	19	53	43	91
Diazinon oxon	13	17	8	17
Dichlorvos	11	0	0	2
Dimethoate	11	1	0	0
Disulfoton	11	0	0	0
Ethion	11	1	0	2
Ethyl parathion	11	1	0	0
Fonofos	12	0	0	0
Malathion	12	18	5	20
Malathion oxon	11	0	0	0
Methamidophos	11	2	1	0
Methidathion	11	1	1	0
Methyl parathion	11	0	0	0
cis-Mevinphos	11	21	7	7
trans-Mevinphos	11	5	0	4
Naled	11	0	0	0
Phosmet	11	2	0	4
OTHER PRODUCTS				
Fipronil	11	8	10	7
Piperonyl butoxide	12	23	11	50

3.0 AIR CONCENTRATION MEASUREMENTS

3.1 Introduction and Data Availability

Children are exposed to residential pesticides via the ingestion, dermal, and inhalation routes. Of these routes, inhalation is the best characterized and requires measurements that are simple to collect in field studies. Estimating absorption via inhalation relies on measured airborne chemical concentrations and on relatively few default exposure factor assumptions, such as the inhalation rate and time spent in specific locations. Since indoor pesticide concentrations are typically higher than outdoor concentrations, and since young children spend the majority of their time indoors, indoor concentrations account for the bulk of their inhalation exposure.

Absorption via the inhalation pathway involves the uptake of vapors and particle-bound residues present in the air. It is generally assumed that inhaled vapors will be readily absorbed across the alveolar membrane into the bloodstream (at least for soluble compounds). Particle-bound residue may vary in size and composition, both of which may influence thoracic penetration and affect absorption. Inhaled particle-bound contaminants trapped in upper airway (nasal and upper lung) mucosa may also be subsequently ingested.

The methods for measuring of airborne pesticide concentrations are well-developed and easily implemented indoors and outdoors using stationary or personal samplers. The methods involve collecting gases and/or particle-bound residues onto filters and sorbent media (the two are combined so that no distinction is made between gases and particle-bound residues). Stationary samplers are typically placed adjacent to treated areas and/or in the location where the participant spends the most time. Samplers may be placed at several locations throughout the home to investigate the spatial distribution of pesticides. Stationary samplers are located at specified heights above the floor to represent the assumed breathing area of the study participants. Personal samplers are worn by the study participants near the breathing zone. Either type of sampler may be modified with a size selective inlet to exclude specific particle size fractions. Sampling media vary but often consist of a pre-filter in tandem with a sorbent composed of polyurethane foam (PUF) or polymeric resin beads (*e.g.,* XAD).

The sampling approaches and methods for each study are described in Table 3.1. Since air sampling techniques are fairly standardized, the methods are consistent across studies. In the large observational field studies, air samples were collected over multiple days for reasons that included reducing measurement error due to day-to-day variability, improving detection limits, and reducing costs associated with changing and analyzing filters. The smaller, focused studies typically employed multiple, consecutive 24-hour sampling periods to capture temporal variability. Personal sampling was attempted in only one study, MNCPES, but compliance issues were noted.

3.2 Pesticide Presence

All pesticides included in this report have been used in residential settings. Because of the potentially long persistence of some pesticides in the indoor environment (Gurunathan *et al.,* 1998), they may be detected even in the absence of a recent application. Detection frequencies for indoor and outdoor samples are presented graphically in Figure 3.1. While detection

frequency corresponds inversely to the limit of detection (LOD), the LOD for each compound is relatively consistent across the large observational field studies. The exception to this is the NHEXAS-Arizona study, which employed a collection method with a relatively small sample volume, resulting in a higher LOD. The LODs for each pesticide by study are presented in Table 3.2.

- Detection limits (Table 3.2) varied by as much as an order of magnitude across studies. Within studies, detection limits were similar for organophosphate and pyrethroid insecticides. Detection limits are influenced by sample volume (Table 3.1). For example, the much lower detection limits for chlorpyrifos and diazinon in MNCPES compared to NHEXAS-AZ reflects the much larger volume sampled in MNCPES.

- The compounds most frequently detected in indoor air (Figure 3.1) were the organophosphate (OP) insecticides chlorpyrifos, (typically > 90%) and diazinon (typically > 75%), followed by the pyrethroid insecticide permethrin (typically > 50%).

- The insecticides most frequently detected in outdoor air (Figure 3.1) were also chlorpyrifos and diazinon, but the detection frequencies were lower and more variable across studies.

- Chlorpyrifos was detected at a high frequency (Figure 3.1) even in those studies conducted after its indoor residential use was restricted (JAX and CHAMACOS).

- The pesticide degradation products of chlorpyrifos and diazinon, TCPy and IMP, respectively, were frequently detected in air samples collected in CTEPP (Figure 3.1); none of the other studies included these as target analytes.

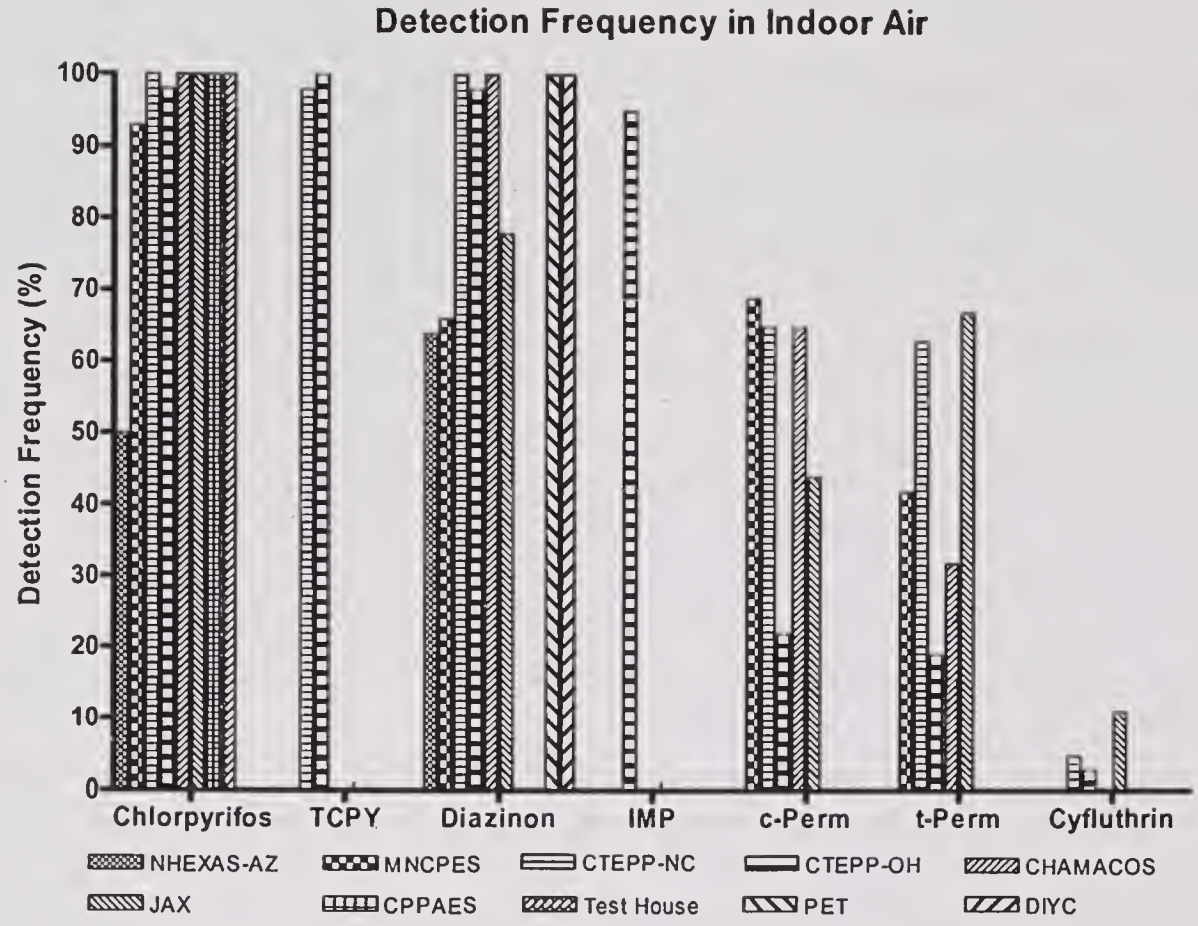
Table 3.1 Summary of air sample collection methods.

Study	Samples Collected	Cohort Size	Sampling Location	Sampling Device	Device Details	Sample Volume	Collection Frequency	Collection After Pesticide Use	Relevant Analytes
NHEXAS-AZ	Indoor	14	Home	Pumps w/ 10 µm inlet, PUF and Teflon-coated glass filters	Intermittent sampling (total of 12 h over 3 d)	Approx 3 m^3 (4 L/min for 12 hr)	Integrated 3-day monitoring period	No	Chlorpyrifos, Diazinon, Malathion
MNCPES	Personal Indoor Outdoor	70 97 52	Home	Pumps w/ XAD cartridge and quartz filter	Backpack carrying case for personal, sound-proof enclosure	Approx 10.8 m^3 (1.25 L/min for 144 hr)	Continuous, Days 1-7, integrated	No	Chlorpyrifos, Diazinon, Malathion, Atrazine
CTEPP	Indoor Outdoor	257	Home and Daycare	Pumps w/ 10 µm inlet, quartz fiber filter and XAD-2 cartridge	Indoor: Styrofoam box w/ cooling fan; Outdoor: plastic dog house. 75 cm height.	Approx 12 m^3 (4 L/min for 48 hr)	One 48-hr sample	No	OPs & Pyrethroids incl. Chlorpyrifos, Diazinon, and Permethrin
JAX 2001	Indoor Outdoor	9	Home	Constant-flow battery powered pump w/ PUF cartridge	Breathing-zone height indoor, 1.5 m height outdoor	Approx 5.5 m^3 (3.8 L/min for 24h)	One 24-hr sample	Yes, indoor	OPs & Pyrethroids incl. Chlorpyrifos, Diazinon, and Permethrin
CHAMACOS	Indoor Outdoor	20	Home	Sampling pump with PUF cartridge	Tamper-resistant box	Approx. 3.6 m^3 (2.5 L/min for 24 hr)	One 24-hr sample	No	OPs & Pyrethroids incl. Chlorpyrifos, Diazinon, and Permethrin
CPPAES	Indoor	10	Home	Harvard Sampler w/ PM_{10} inlet, cotton filter impregnated w/ activated carbon	Placed in room most frequented by child, approx 1 m high.	Approx.14 m^3 (24h) and 29 m^3 (48h)	Four 24-hr samples on days 0-3; four 48-hr samples days 3-11	Yes, indoor	Chlorpyrifos
Test House	Indoor	1	Test House	Low volume pump w/PUF	Multiple rooms	Approx 5 m^3 (3.5 L/min for 24 hr)	Time series over 21 days	Yes	Chlorpyrifos
PET Pilot Study	Indoor	6	Home	Low volume pump w/PUF	Living room and child's bedroom	Approx 5 m^3 (3.5 L/min for 24 hr)	24-hr samples: Pre-application and days 1, 2, 4, & 8 post-application	Yes, lawn application	Diazinon
DIYC	Indoor Outdoor	3	Home	Pump w/XAD	Placed in room most frequented by child,	Approx. 11.5 m^3 (8 L/min for 24 hr)	One pre- and six post-application measurements	Yes, indoor (2 professional, 1 resident)	Diazinon

Table 3.2 Limits of detection (ng/m³) for air samples by compound and study.

Compound	Chlorpyrifos	Diazinon	*cis*-Permethrin	*trans*-Permethrin	Cyfluthrin	TCPy	IMP
NHEXAS-AZ	3.2	2.1	--[a]	--	--	--	--
MNCPES	0.10	0.10	0.09	0.09	--	--	--
CTEPP NC	0.09	0.09	0.09	0.09	0.87	0.09	0.09
CTEPP OH	0.09	0.09	0.39	0.33	0.87	0.09	0.09
JAX	1.0	0.4	1.0	1.0	1.2	--	--
CHAMACOS	0.3	0.3	0.6	0.6	7.0	--	--
CPPAES	2.0	--	--	--	--	--	--
DIYC	--	1.2	--	--	--	--	--
PET	--	1.0	--	--	--	--	--

[a] Blank cells (--) indicate that the pesticide or metabolite was not measured in the study.



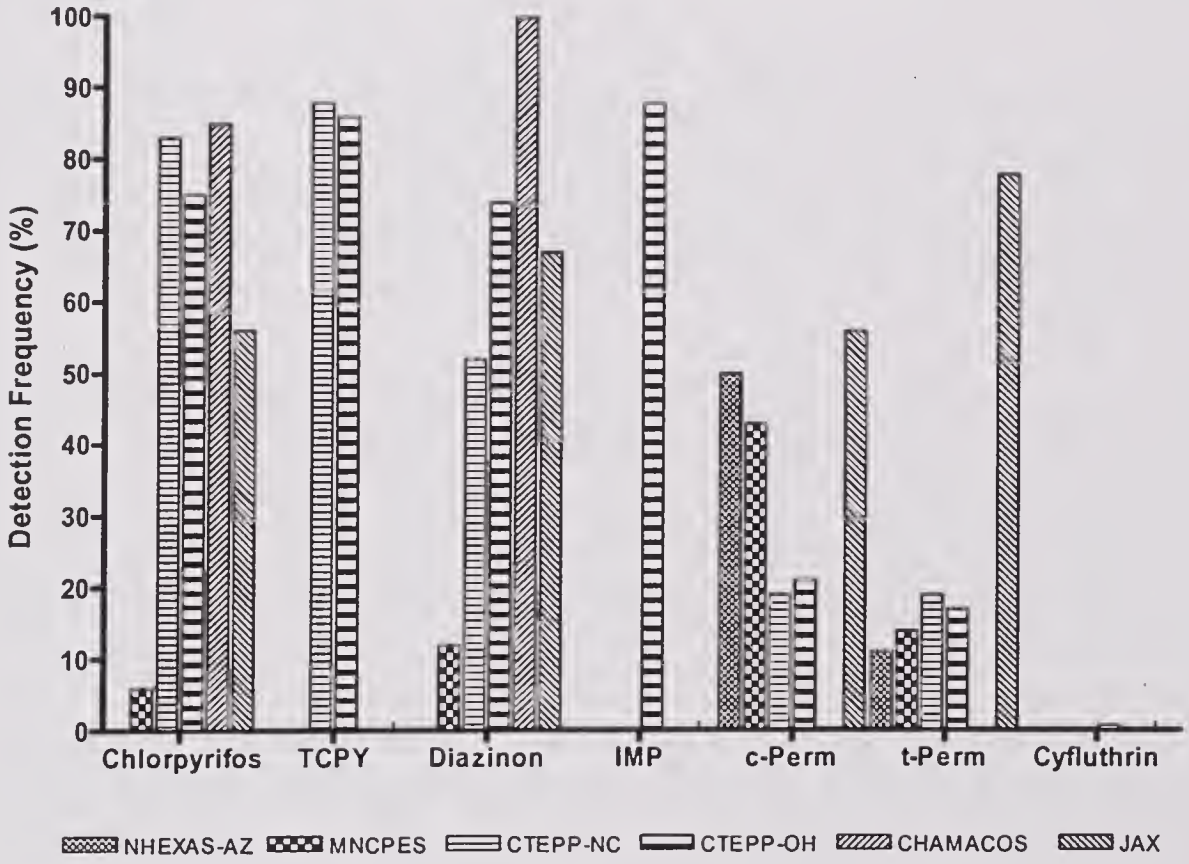


Figure 3.1 Frequency of detection of pesticides measured in indoor and outdoor air in selected studies.

3.3 Comparisons of Air Concentrations

Previous studies have reported post-application concentrations of semi-volatile pesticides in air that may reach levels representing considerable exposure by the inhalation route (Byrne *et al.,* 1998; Fenske *et al.,* 1990; Lewis *et al.,* 2001). Low measurable airborne levels have also been reported even in the absence of a recent application event (Lewis *et al.,* 1994; Whitmore *et al.,* 1994). Lognormal probability plots and box-and-whisker plots graphically depicting the (unweighted) measurements of compounds of interest in our studies are presented in Figures 3.2 through 3.5. The median and 95th percentile concentrations are presented in Table 3.3 (complete summary statistics are presented in Tables A.1 through A.7 in Appendix A).

- For pesticides measured in indoor and outdoor air, the observed concentrations typically approximate lognormal distributions, as demonstrated in the lognormal probability plots in Figures 3.2 and 3.3.

- Despite differences in the lengths of the sample collection periods (1 to 7 days), the in-door chlorpyrifos concentrations observed across the large observational field studies are similar in their variability, as demonstrated by similar slopes in the probability plot (Figure 3.2). Similar variability over varying collection periods suggests that air concentrations are reasonably consistent from day-to-day in the absence of a recent application.

- Comparison of air concentrations across studies in the box-and-whisker plots (Figure 3.4) finds that, as expected, pesticide concentrations in smaller studies, where measurements immediately followed an application, are much higher than in the larger observational field studies; for example, note the high indoor chlorpyrifos levels measured in CPPAES and the Test House.

- Median concentrations are typically an order of magnitude higher indoors than outdoors (Table 3.3). Two notable exceptions are JAX and CHAMACOS. In the JAX samples, collected in a community with high year-round pesticide usage, outdoor diazinon and *cis*- and *trans*-permethrin levels are nearly as high as indoor levels. In the CHAMACOS samples, collected in an agricultural community, median outdoor diazinon levels exceed indoor levels.

- The low pesticide concentrations routinely measured outdoors (notwithstanding the exceptions noted above) together with the relatively short amount of time that young children typically spend outdoors suggest that inhalation of outdoor air is not an important contributor to their aggregate pesticide exposure.

- The median indoor concentrations in the large observational field studies are higher for the organophosphates (OPs) than for the pyrethroids (Figure 3.4). Not only do OPs tend to have higher vapor pressure, but at the time these studies were conducted, OPs still dominated the marketplace. Detectable levels of chlorpyrifos and diazinon are likely to exist for some time after restriction of their indoor uses due continued use of existing home inventories and reemission from indoor surfaces serving as sinks (such as carpet).

- In indoor air measured in CTEPP (Figure 3.6), a relationship is evident between chlorpyrifos and its degradation product TCPy. The same is true for diazinon and its degradation product IMP. The nearly log-log relationship suggests a power relationship, and at the median level the degradate is present at about 25 to 30% of the concentration of its parent. Accordingly, the metabolites/degradates measured in urine may reflect exposure to both the parent pesticide and the degradate, not just to the parent compound as is often assumed.

- Environmental concentrations of the degradation products were not measured in any of the small, pilot-scale studies, thus the degradate-to-parent ratio immediately following application is unknown.

Table 3.3 Median and 95th percentile air concentrations (ng/m^3, unweighted) for frequently detected pesticides.

		Chlorpyrifos		Diazinon		*cis*-Permethrin		*trans*-Permethrin	
Study	Location	P50	P95	P50	P95	P50	P95	P50	P95
NHEXAS-AZ	Indoor	3.37	164.7	5.59	219.6	--[a]	--	--	--
	Outdoor	ND[b]	ND	ND	ND	--	--	--	--
MNCPES	Personal	1.52	16.86	0.28	4.66	0.20	2.07	<0.09	1.72
	Indoor	1.85	30.25	0.27	8.59	0.09	1.26	<0.09	1.26
	Outdoor	<0.10	0.19	<0.10	0.22	<0.09	0.15	<0.09	0.48
CTEPP-OH[c]	Indoor	1.75	21.69	0.97	56.87	0.28	1.63	0.23	1.04
	Outdoor	0.20	1.13	0.15	1.49	0.28	0.95	0.23	0.66
CTEPP-NC[c]	Indoor	6.07	62.22	2.03	63.66	0.41	7.79	0.27	7.16
	Outdoor	0.28	3.99	0.09	0.98	0.06	0.47	0.06	0.30
JAX	Indoor	20.37	84.92	4.64	28.04	0.71	92.47	3.06	134.3
	Outdoor	3.77	6.62	3.53	6.76	2.13	2.29	2.50	10.24
CHAMACOS	Indoor	1.90	NA[d]	1.80	NA[d]	0.50	NA[d]	<0.10	NA[d]
	Outdoor	0.90	NA[d]	2.80	NA[d]	0.10	NA[d]	<0.10	NA[d]
CPPAES[e]	Indoor	149.0	815.6	4.55	23.88	--	--	--	--
Test House[e]	Indoor	290.0	1000	--	--	--	--	--	--
PET	Indoor	--	--	45.6	562	--	--	--	--
DIYC	Indoor	--	--	1800	4900	--	--	--	--

[a] Blank cells indicate the pesticide was not measured in the study
[b] ND = not detected
[c] CTEPP samples collected at both homes and daycares
[d] NA = summary statistic not available at time the report was prepared
[e] Day 1 measurements only, multiple rooms

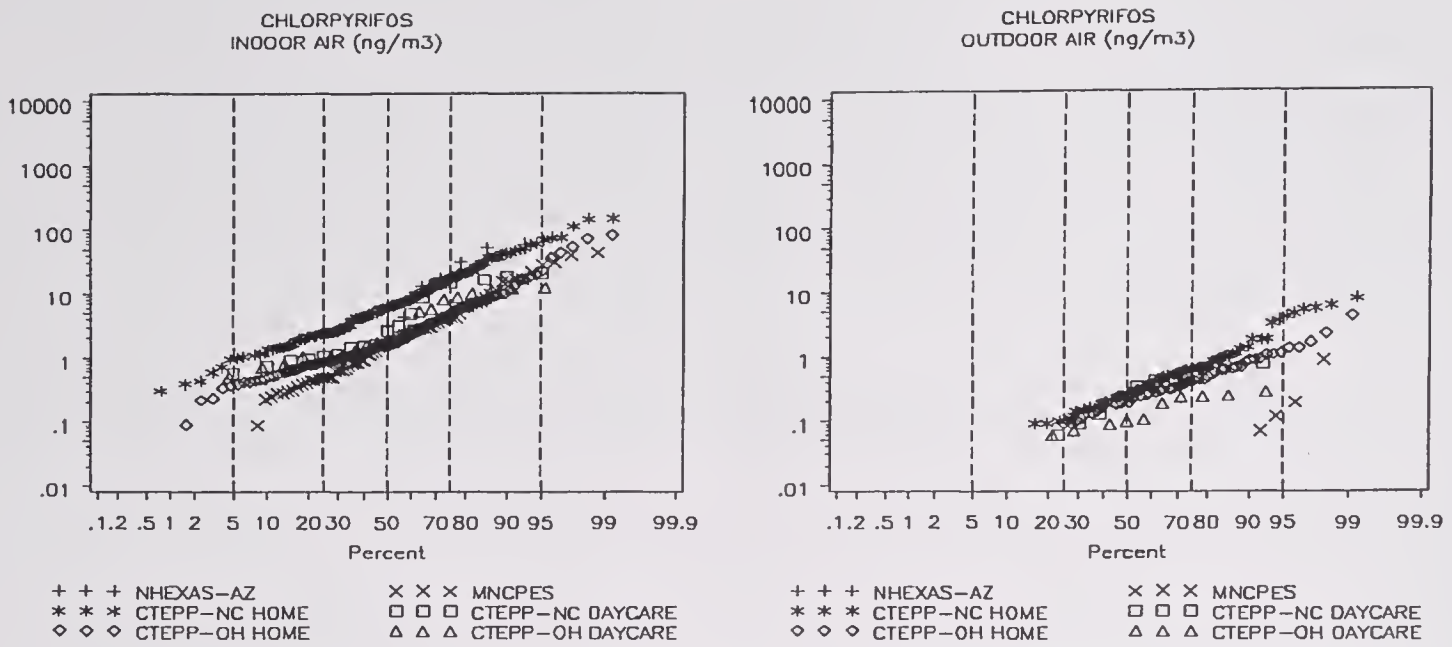


Figure 3.2 Log probability plots for chlorpyrifos, diazinon, and *cis*-permethrin measured in large observational field studies. Only values above the limit of detection are plotted.

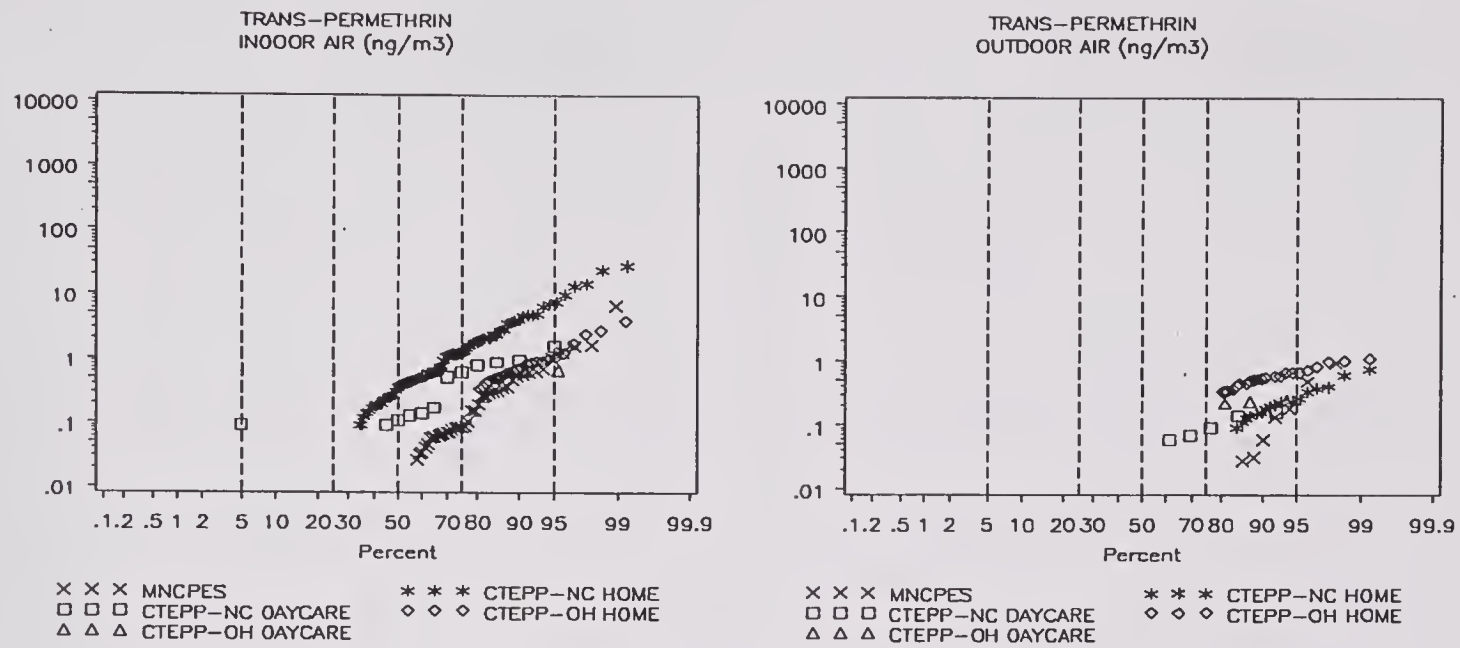


Figure 3.3 Log probability plots for *trans*-permethrin, TCPy, and IMP measured in large observational field studies. Only values above the limit of detection are plotted.

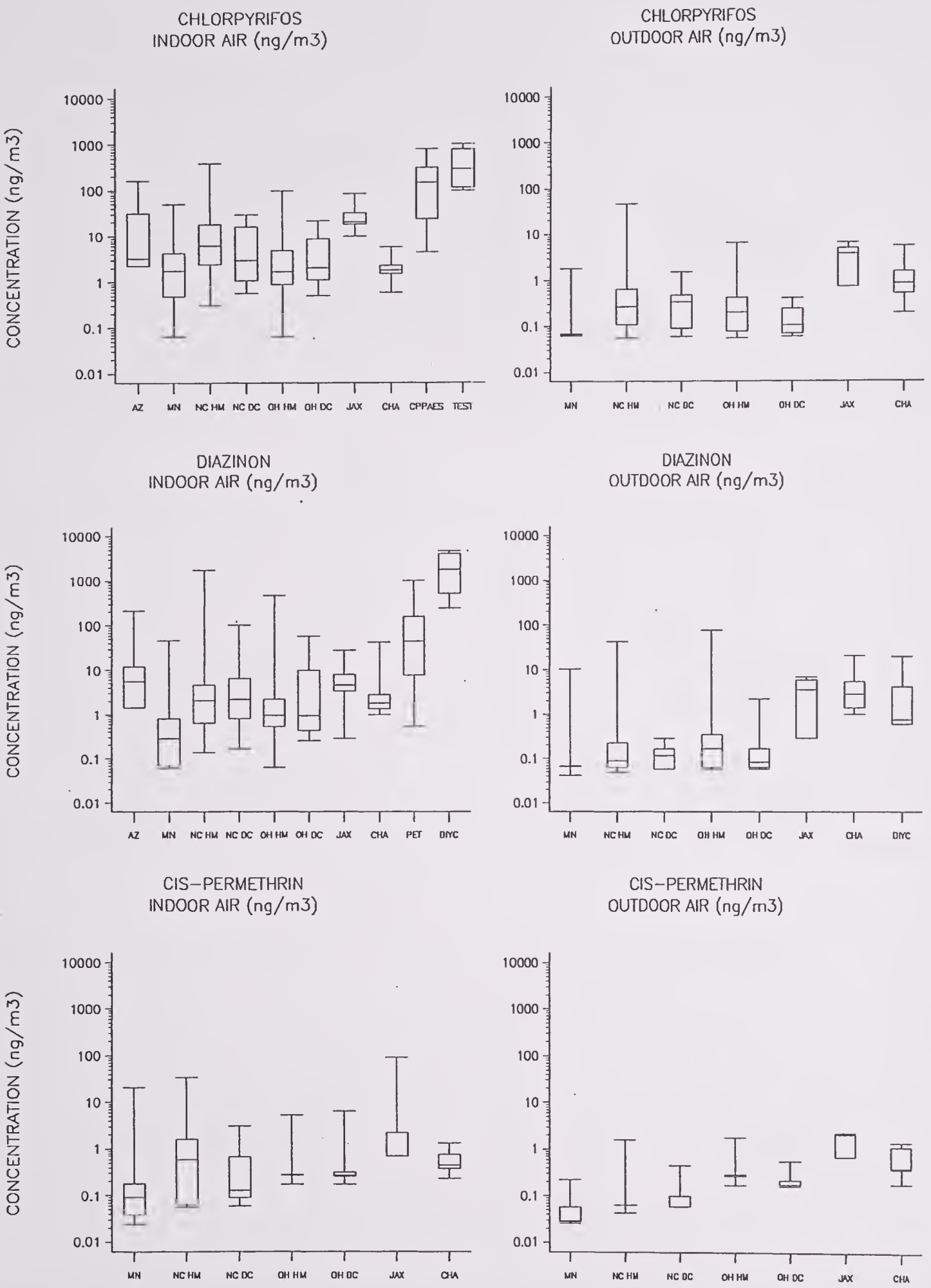


Figure 3.4 Indoor and outdoor air concentrations of chlorpyrifos, diazinon, and *cis*-permethrin measured in selected studies. Legend: AZ = NHEXAS-AZ, MN = MNCPES, NC HM = CTEPP-NC Home, NC DC = CTEPP-NC Daycare, OH HM = CTEPP-OH Home, OH DC = CTEPP-OH Daycare, CHA = CHAMACOS, TEST = Test House.

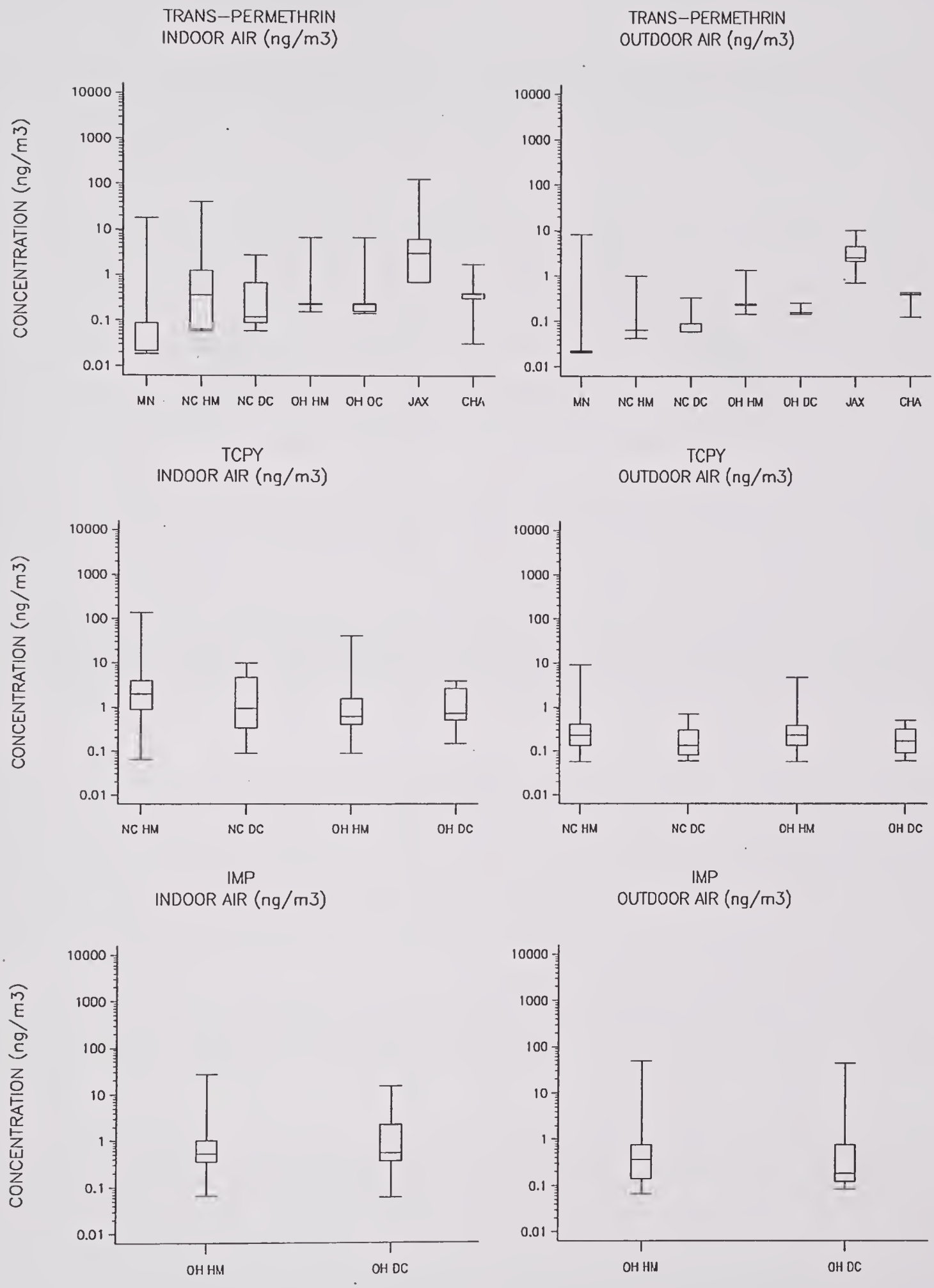


Figure 3.5 Indoor and outdoor air concentrations of *trans*-permethrin and TCPy measured in selected studies. Legend: AZ = NHEXAS-AZ, MN = MNCPES, NC HM = CTEPP-NC Home, NC DC = CTEPP-NC Daycare, OH HM = CTEPP-OH Home, OH DC = CTEPP-OH Daycare, CHA = CHAMACOS, TEST = Test House.

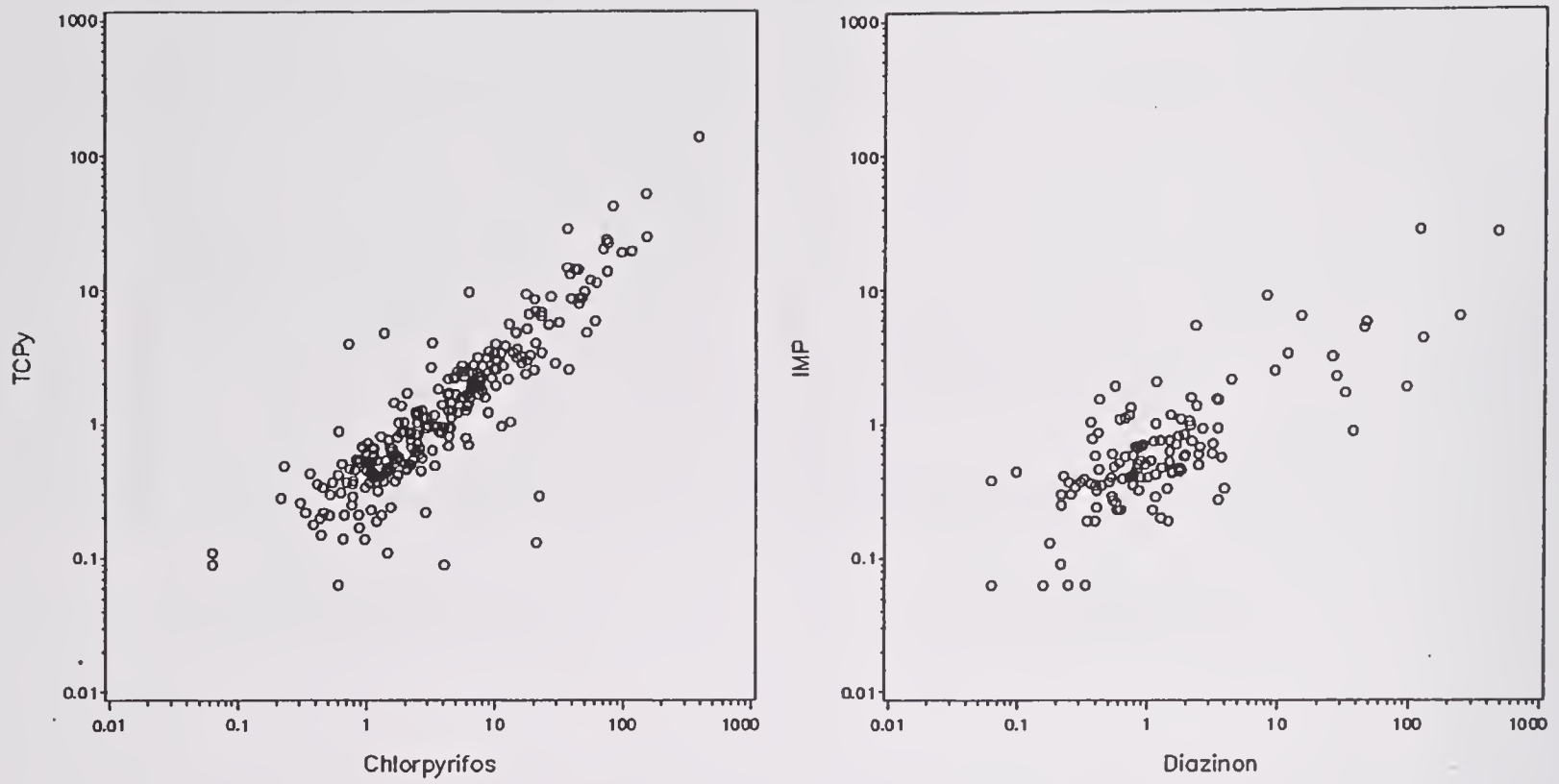


Figure 3.6 Log-scale relationships between levels of parent pesticide (ng/m^3) and degradate (ng/m^3) measured in CTEPP. Left Panel: Chlorpyrifos with TCPy. Right Panel: Diazinon with IMP.

3.4 Differences Related to Location

This section addresses differences in potential for exposure related to geographic region, population density (urban *vs.* rural), and home *vs.* daycare environment. There is available evidence to support all three of these location-related factors as having a discernable impact on pesticide exposure.

The large observational field studies were conducted in several geographical regions. A difference in climate impacts the type and density of pests found in the region. Residents of areas with mild winter conditions, as exist in the southern United States, may experience significant pest control problems throughout the year and may respond with increased pesticide usage. The landmark EPA Non-Occupational Pesticide Exposure Study (NOPES) conducted during 1986-1988 (Whitmore *et al.,* 1994) reported much higher indoor air concentrations of chlorpyrifos and diazinon in Jacksonville, Florida, than in Springfield and Chicopee, Massachusetts (purposely selected as high-use and low-use regions, respectively).

The residents of rural communities may be exposed to pesticides from residential as well as agricultural applications. Both spray drift and work-to-home transport are potential pathways of exposure to agricultural pesticides, some of which have the same active ingredient as formulations used within the home (Curl *et al.,* 2002). Residents of urban areas, on the other hand, may experience frequent applications to combat persistent pest control problems arising from high population density (Landrigan *et al.,* 1999), may have little control over pesticide applications by building management, and may be exposed to pesticides applied in neighboring residences.

Young children spend nearly 20 hours per day indoors (US EPA, 2002). For pre-school age children, much of this time is spent in residences or in daycare facilities. According to recent estimates, nearly 4 million children under age 6 spend some portion of their day in center-based child care, with many children spending a full work day (8-10 hours) in the child care center (US CPSC, 1999). Pesticide concentrations in daycare facilities are potentially significant (Wilson *et al.,* 2003) and are typically out of the control of the parents.

- Positive and highly significant associations ($p < 0.01$) between personal-air exposures and indoor air concentrations were observed in MNCPES for both chlorpyrifos and diazinon with Spearman correlation coefficients of 0.81 and 0.62, respectively (Table 3.4).

- Comparison of the box-and-whisker plots in Figure 3.4 of indoor air concentrations measured in homes finds median values were somewhat higher in southern states (NHEXAS-AZ and CTEPP-NC) than in northern states (MNCPES and CTEPP-OH). However, considerable overlap in the interquartile ranges is evident. Since these studies focus on compounds that have been used to control a variety of common insect pests both inside and outside of homes (chlorpyrifos was until recently among the most poplar residential insecticides for cockroach, flea, ant and termite control), it is not surprising that the distributions would overlap across geographical locations.

- When daycare measurements are included, a geographical difference is less obvious (results not shown). Despite recent gains in the adoption of integrated pest management policies, many daycare facilities still have regular calendar-based pesticide treatments, irrespective of actual demonstrated need. This may have the effect of minimizing differences in usage in daycares among geographic regions.

- CTEPP data (Figure 3.7) suggest that, within each state, indoor air levels in daycares are similar to those in homes, particularly for diazinon and permethrin. This demonstrates the potential for continued exposure as a child transitions from the home to a daycare. To reduce the uncertainty of risk assessments for children, their exposures must be considered for all indoor and outdoor environments they occupy, including homes, child care centers, and other buildings. Additional information may be required to examine exposure potential from schools, restaurants, and other public and private locations where pesticides are also applied.

- Differences between urban and rural air concentrations of chlorpyrifos were observed in both MNCPES (Table 3.5) and CTEPP-OH (Table 3.6). The differences reached statistical significance only in MNCPES, with higher concentrations in the urban areas. Likewise, the detection frequencies for both chlorpyrifos and diazinon in indoor and personal air were higher in urban locations (Table 3.5).

- Across compounds in MNCPES, median levels were consistently higher in urban areas than in rural areas. A reasonable explanation may be that urban areas require more intensive use of pesticide products to control a range of pests over a wider seasonal span. In addition the application may be of more mass of active ingredients in a smaller area, as is the case with a liquid termiticide application. While it is not entirely clear why the pattern of higher urban levels was not evident in CTEPP-NC, it may be due to a less stringent definition of "urban" in CTEPP.

- Air samples collected in low-income homes generally had higher concentrations of chlorpyrifos and diazinon than samples collected in medium/high income homes (Table 3.6), but the difference was only statistically significant for diazinon in NC.

Table 3.4 Spearman correlations among personal, indoor, and outdoor concentrations of chlorpyrifos and diazinon measured in MNCPES[a].

	Chlorpyrifos		Diazinon	
Type	Indoor	Outdoor	Indoor	Outdoor
Personal	0.81**	0.23	0.62**	0.67**
Indoor	--	-0.01	--	0.28

[a] Excerpted from Clayton *et al.*, 2003
** Statistically significant at the 0.01 level.

Table 3.5 Urban and rural differences in airborne concentrations of chlorpyrifos and diazinon measured in MNCPES. The limit of detection was 0.1 ng/m^3.

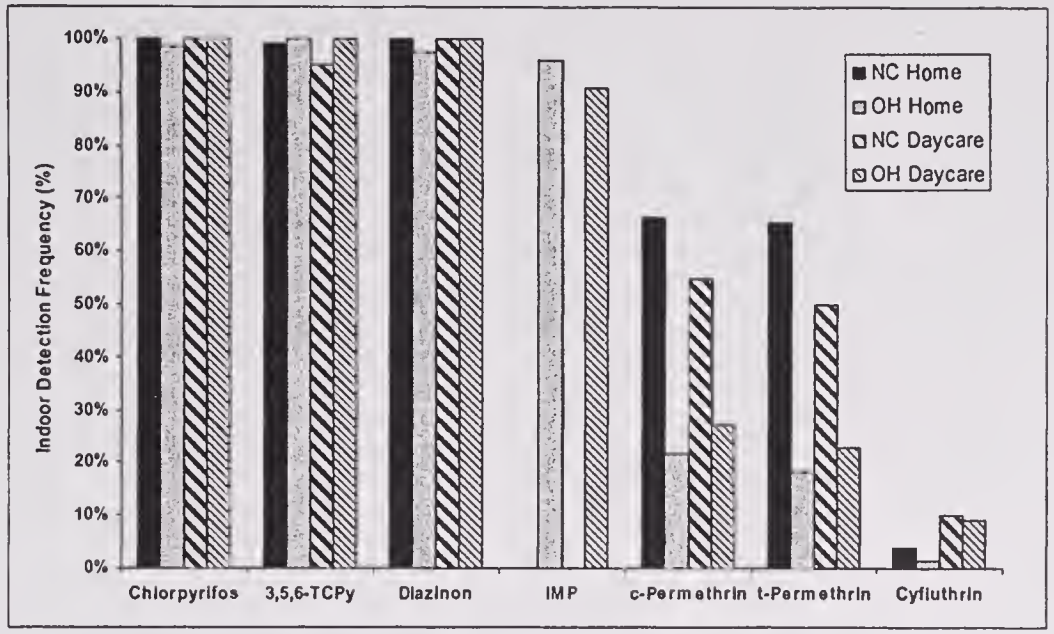
Sample Type	Chemical	Location	N	Detection Frequency	Median Concentration (ng/m^3)
Personal	Chlorpyrifos*	Urban/Suburban	40	98%	2.2
		Rural	20	90%	1.2
	Diazinon*	Urban/Suburban	30	77%	0.4
		Rural	18	44%	<0.1
Indoor	Chlorpyrifos*	Urban/Suburban	57	96%	2.2
		Rural	25	80%	0.7
	Diazinon	Urban/Suburban	54	74%	0.4
		Rural	21	52%	0.1

* denotes significant ($p < 0.05$) difference in medians using two-sided Wilcoxon test.

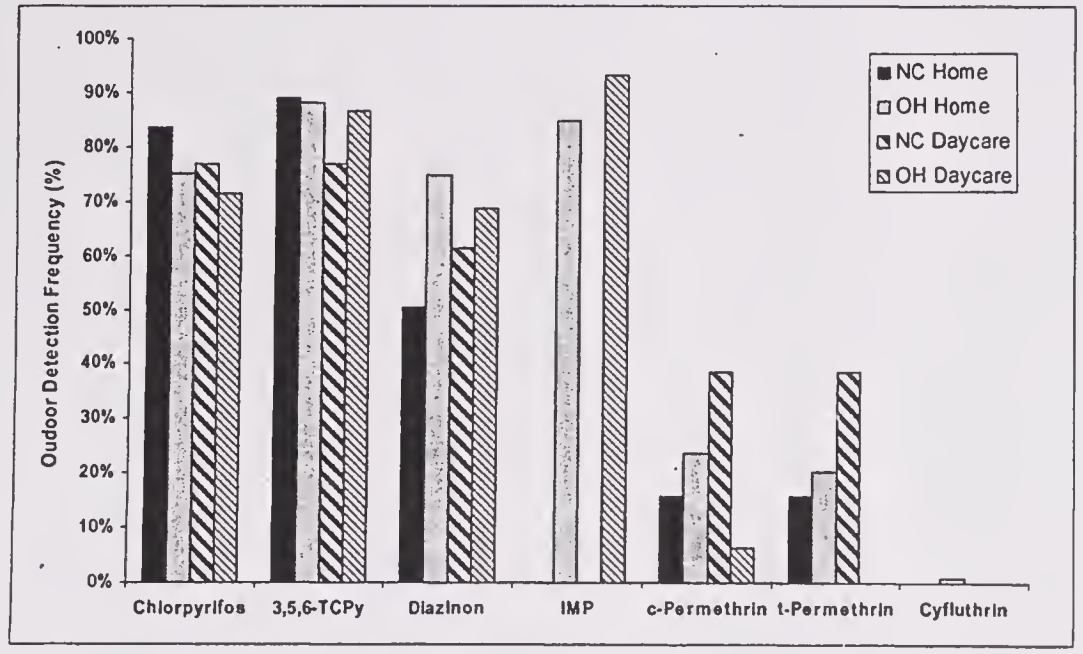
Table 3.6 Differences in airborne concentrations measured in CTEPP for urban versus rural, low versus medium income, and home versus daycare expressed as ratios of geometric means. Adapted from Morgan *et al.*, 2004.

State	Chemical	Estimated Ratio of Geometric Means (95% C.I.)		
		Urban/Rural	Low /Mid-High Income	Home/Daycare
North Carolina	Chlorpyrifos	0.94 (0.50, 1.77)	1.36 (0.84, 2.21)	1.78 (0.81, 3.92)
	Diazinon	0.95 (0.43, 2.11)	3.59* (1.95, 6.61)	0.82 (0.30, 2.24)
Ohio	Chlorpyrifos	1.64 (0.80, 3.37)	1.63 (0.97, 2.74)	0.76 (0.38, 1.52)
	Diazinon	1.04 (0.44, 2.49)	1.67 (0.89, 3.12)	0.78 (0.34, 1.80)

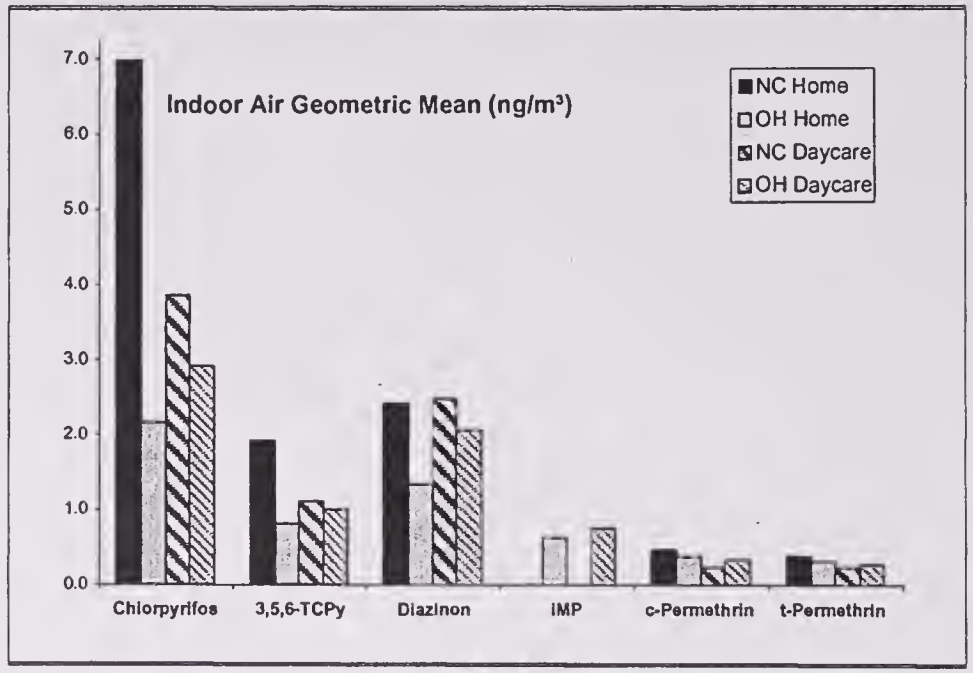
* denotes significance, $p < 0.05$.



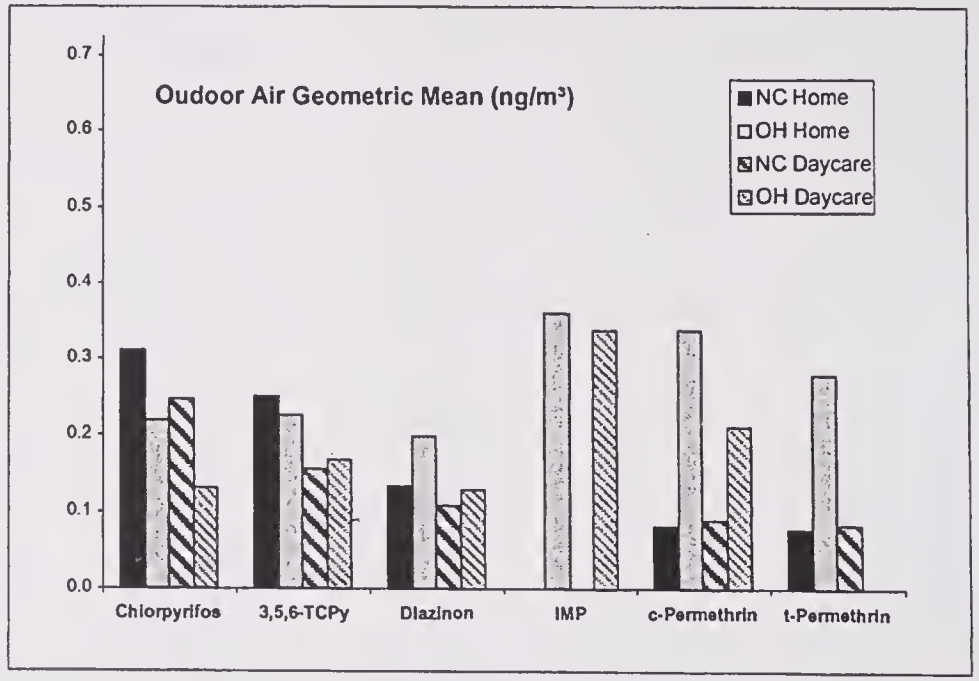
A



B



C



D

Figure 3.7 The detection frequencies of select pesticides and their metabolites measured from the indoor air (A) and outdoor air (B) of homes and daycares in NC and OH, and the mean concentrations of select pesticides and their degradation products measured from the indoor air (C) and outdoor air (D) of homes and daycares in NC and OH.

3.5 Spatial and Temporal Variability

Few studies have been designed to measure either the spatial variability of airborne pesticide concentrations in a home or the temporal variability following crack-and-crevice pesticide applications (Byrne *et al.,* 1998; Lewis *et al.,* 2001). Recently, the Test House, CPPAES, DIYC, and PET studies have provided data on both spatial and temporal variability, as shown in Figure 3.8.

- Within-home spatial patterns were investigated in the Test House experiments. Following a crack and crevice application of chlorpyrifos (Figure 3.8 and Table 3.7), the pesticide was detected in the application room (kitchen), adjacent den, and the farthest bedroom from the application. Airborne concentrations in the kitchen peaked at 790 ng/m^3, then decreased by approximately 80%, but were still measurable, at 21 days after application. A concentration gradient was observed from the kitchen (application area) to the den (proximal area) to the master bedroom (distal area).

- Between-home spatial variability following a pesticide application was investigated in the CPPAES and DIYC studies. Indoor air concentrations of chlorpyrifos among the 10 homes in the CPPAES spanned more than an order of magnitude one day after application (Figure 3.8).

- The highest measured chlorpyrifos indoor air concentrations following crack and crevice applications among a subset of 5 CPPAES homes were between days 0 and 2 post application (mean = 315 ng/m^3), then decreased throughout the 2-week sampling period (mean = 172 ng/m3), but were still greater than the pre application levels (mean = 18 ng/m^3). The indoor air concentrations for the remaining CPPAES homes were much lower and did not follow the same decay pattern (data not presented, see Hore *et al.,* 2005).

- Air concentrations of diazinon in the homes of the DIYC study were nearly an order of magnitude higher than concentrations of chlorpyrifos in CPPAES, and the decay pattern differed dramatically among the three DIYC homes. The difference in airborne diazinon concentrations among the three homes was most pronounced 4-5 days after application (Figure 3.8), perhaps partially attributable to both the application method employed and the amount of active ingredient applied in each home.

- Following outdoor granular application to lawns in the PET study, indoor air concentrations of diazinon generally reached maximal levels by days 1 and 2 post application and declined over the duration of the study (Figure 3.8).

3.6 Factors that Influence Air Concentrations

Multiple factors influence the concentration of pesticides in air and the potential for inhalation exposure. The physico-chemical characteristics of the chemicals applied, the formulation type and the frequency of application are believed to be some of the most important of these factors. Other factors such as seasonal variation, housing type, pets, occupancy, application location, type of surface to which the applications are made, and the rooms where the samples are collected may also influence the concentrations measured. Some of these factors have been

investigated using the data from NERL's pesticide exposure measurement program.

- The impact of air exchange rate (AER) on air concentrations is shown in Figure 3.8 for the CPPAES data. Indoor air concentrations of chlorpyrifos (immediately following application) among the homes spanned more than an order of magnitude. Homes with low air exchange rates had higher initial airborne concentrations and a noticeably slower reduction of airborne levels.

- The amount, or mass, of active ingredient applied also clearly affected the concentrations measured in CPPAES, with low airborne concentrations observed in three homes receiving applications containing only trace amounts of chlorpyrifos (data not presented, please see Hore *et al.,* 2005).

- An empirically derived Application Effective Volume (AEV, applied mass divided by the product of air changes per hour and home volume) was applied to the CPPAES data to demonstrate the relationship between measured air concentrations, air exchange rate, and mass of active ingredient applied. Measured airborne concentration was more consistently correlated with AEV than with any of the constituents of AEV (Pearson product-moment correlations, data not presented). The association of AEV with airborne concentrations measured on the second day after application (Figure 3.9) suggests that AEV may serve as an effective surrogate for air concentrations and that constituent measures including air exchange rate are important determinants of air concentrations.

- The geometric mean concentrations of the organochlorine, organophosphate, and pyrethroid pesticides measured in indoor air in the absence of a recent application appear to be strongly influenced by vapor pressure. Regressing concentrations measured in the CTEPP study upon the logged vapor pressures (Figure 3.10) results in nearly equivalent R^2 values of 0.69 and 0.70 for homes and daycares, respectively. The importance of inhalation as a route of exposure for pesticides is likely to decrease as less volatile pesticides are introduced into the market.

- Results in the US EPA Research Test House comparing total release aerosol to crack and crevice applications confirm that the application method is an important factor influencing the measured airborne concentration of chlorpyrifos (Table 3.7). The application method is also suspected of being a factor responsible for the differences observed among homes in the DIYC study.

- The PET study demonstrates the intrusion of diazinon from an outdoor source. The lawn applications resulted in a source of diazinon that contributed to indoor concentrations in all homes. Indoor concentrations are likely associated with both the physical translocation of particle bound residues and the intrusion of volatilized diazinon from the source. The results suggest that lawn applications increase the potential for occupant exposure both on the treated lawns and indoors.

- While some progress has been made in understanding the multitude of factors that influence the concentration of pesticides in air and the potential for inhalation exposure, additional studies are needed.

3.7 Summary: Air Concentrations

As shown in the bulleted lists of observations from these studies, there are a number of factors that may impact children's exposure to pesticides in homes and child care centers. They include the following:

- The physical and chemical characteristics of the pesticides used indoors will have a significant impact on exposure via the inhalation route. Airborne concentrations will be higher for the more volatile pesticides, such as chlorpyrifos and diazinon (no longer registered for indoor use). Use of less volatile alternatives, such as the pyrethroids, will likely result in lower airborne concentrations of the active ingredients.

- The type and method of pesticide application (see Section 2.4) are factors affecting exposure. As shown in the Test House experiments, the airborne concentrations are higher for foggers than for crack and crevice applications. Past studies have focused on crack and crevice and other spray applications, although newer types of applications, such as use of gels, may further reduce the translocation of pesticides to areas that may be contacted by children.

- The data from these studies highlight the importance of geographic location on airborne concentrations. Frequency of application and total amount of pesticide used may be associated with geographic location.

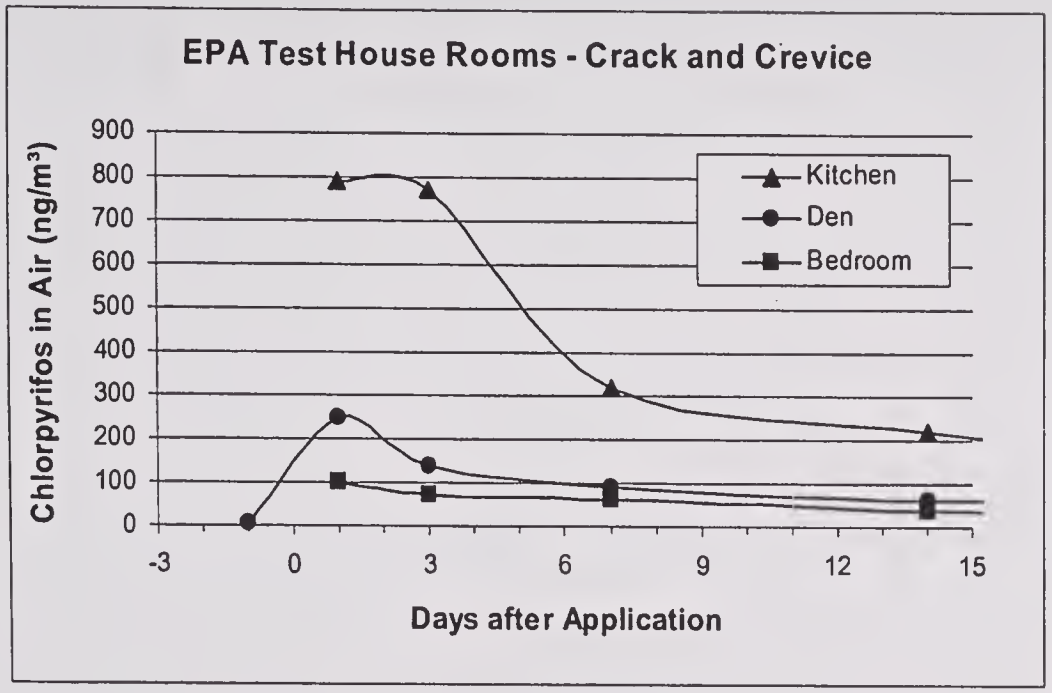
- The data on spatial variability of pesticide residues within a home are limited. But, data from the Test House and other studies show that pesticides are distributed to other locations within a building from the point of application and are measurable in air samples collected in other rooms.

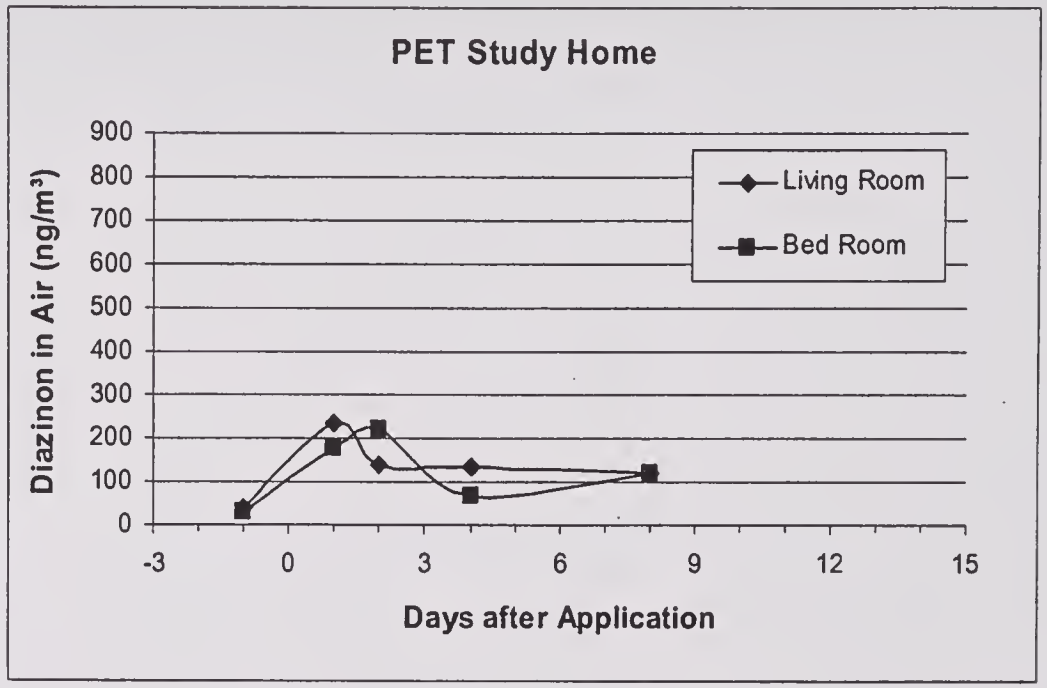
- The data also clearly show that there are temporal changes in concentrations following an application. These changes are related to air infiltration and air exchange rates in the home. The changes are also likely related to degradation processes, but there are few studies that have addressed the temporal changes in concentration for different pesticides as related specifically to the degradation process.

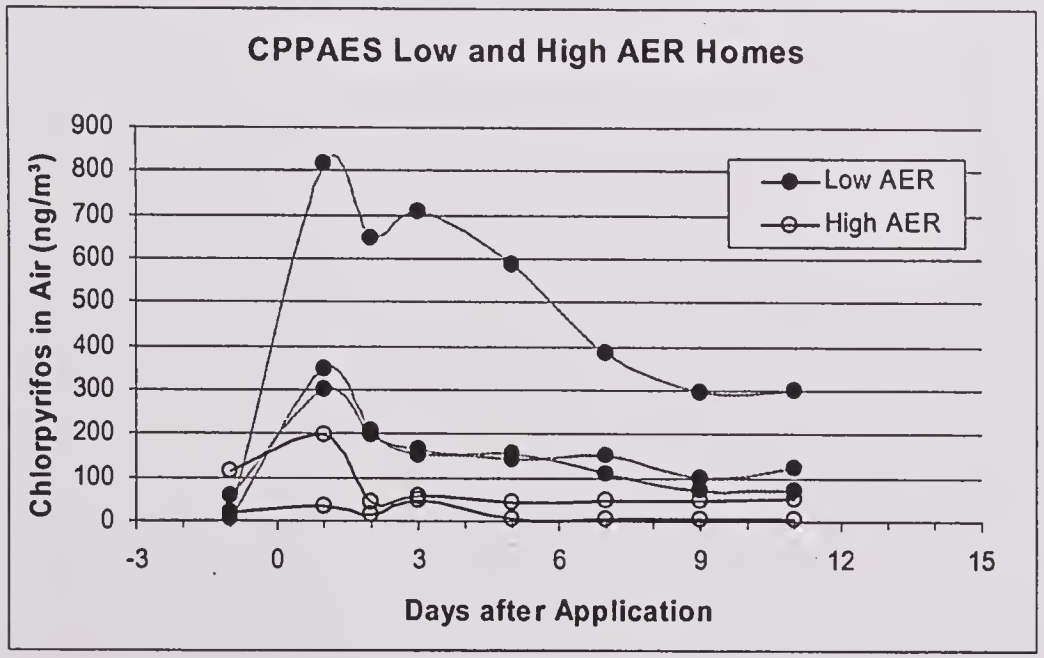
Table 3.7 Airborne chlorpyrifos residues collected following a crack and crevice type application versus a total release aerosol in the EPA Test House.

Application Type	Room	Indoor Air Concentration (ng/m³)							
		Pre	3 hr	Day 1 [a]	Day 2	Day 3	Day 7	Day 14	Day 21
Crack and Crevice	Kitchen	NC [b]	NC	790	NC	770	320	220	140
	Den	3	NC	250	NC	140	90	60	70
	Bedroom	NC	NC	100	NC	0.07	60	40	30
Total Release Aerosol	Living Room	ND [c]	15	9200	4100	2300	860	450	NC
	Den	ND	17	8300	4000	2100	1100	410	NC
	Bedroom	NC	1.4	4700	NC	NC	370	320	NC

[a] Air sampling was initiated immediately following the application and monitored continuously for 24-h.
[b] NC indicates the sample was not collected.
[c] ND indicates the sample was not detected <0.05 μg/m³







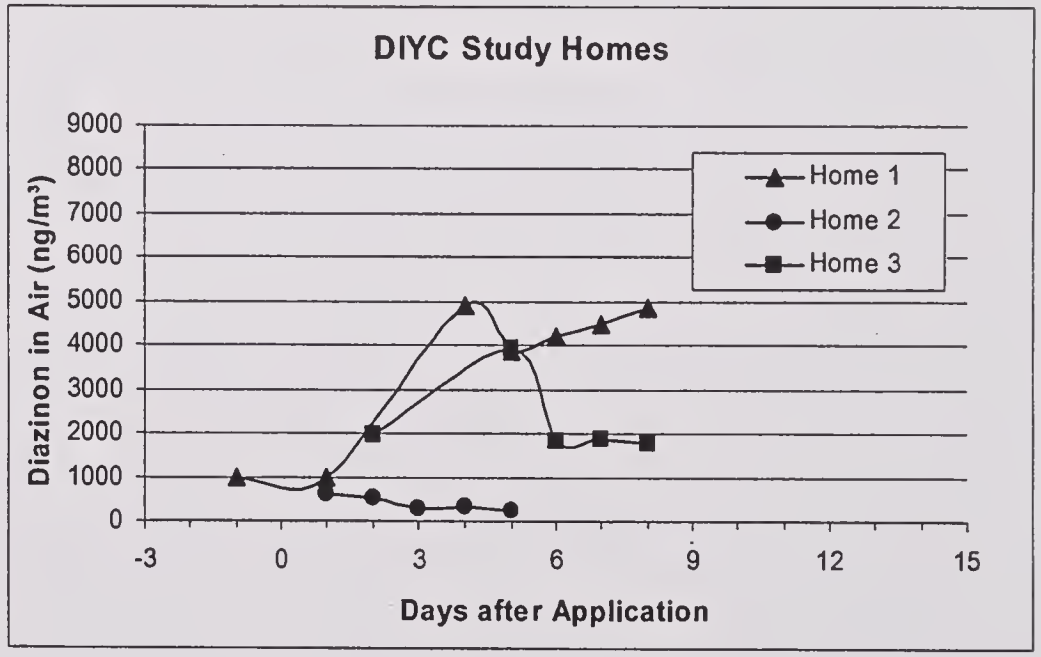


Figure 3.8 Airborne concentrations (ng/m³) of chlorpyrifos or diazinon measured from indoor air over time in the Test House, PET, CPPAES, and DIYC studies.

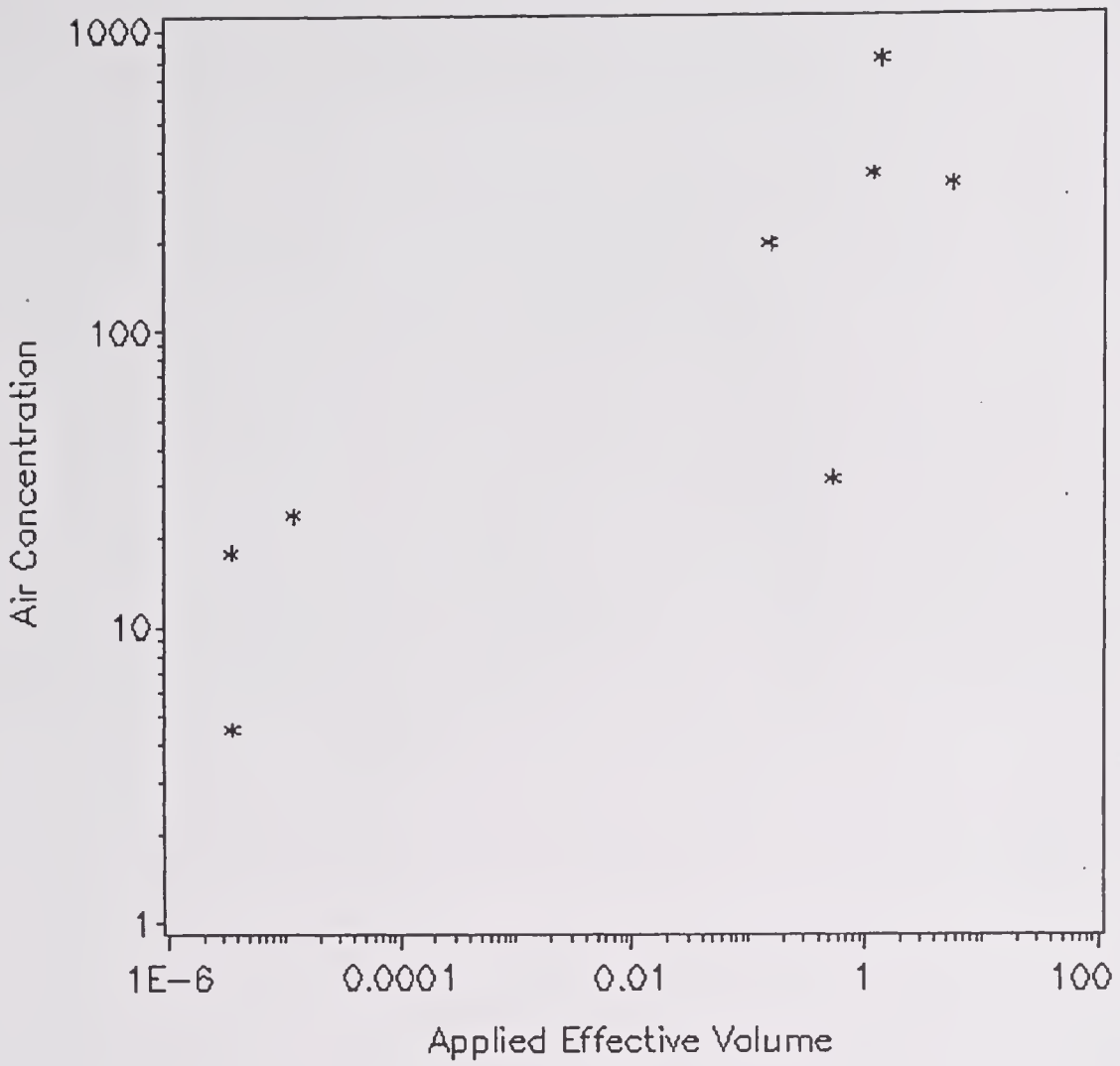


Figure 3.9 Association between measured air concentration (ng/m³) and Applied Effective Volume (ng/m³/h) on the second day after application of chlorpyrifos in CPPAES homes.

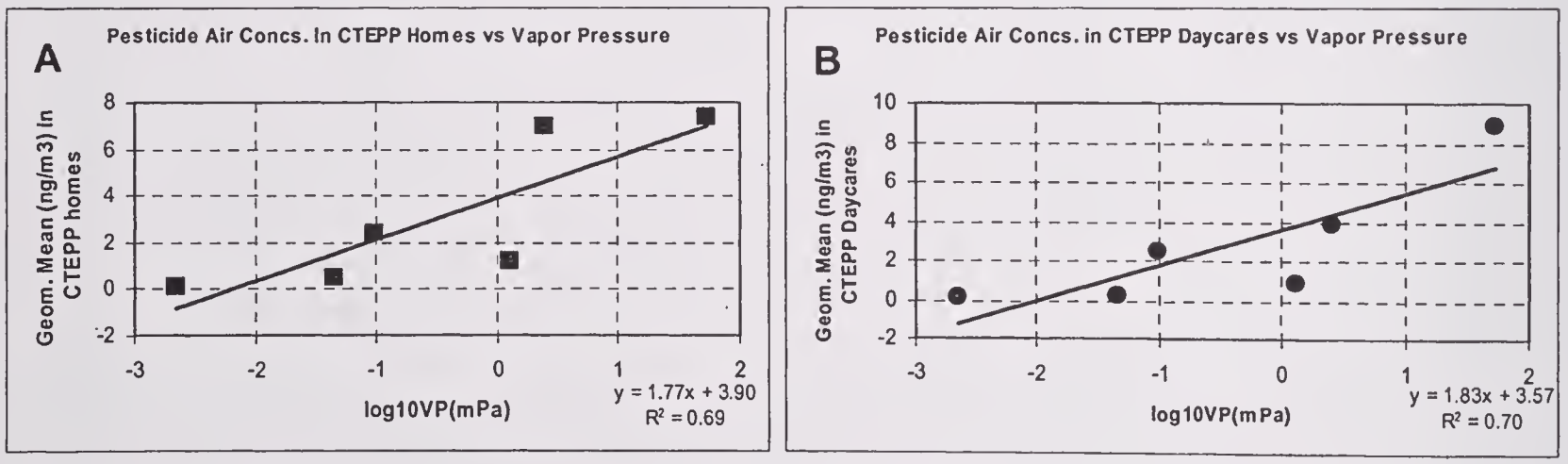


Figure 3.10 Pesticide air concentrations as a function of vapor pressure in CTEPP homes (A) and daycares (B).

4.0 SURFACE MEASUREMENTS

4.1 Introduction and Data Availability

The objectives of measuring pesticide surface residue concentrations and loadings are to describe the extent and distribution of concentrations, identify possible sources of indoor contamination, evaluate factors that may impact concentrations, and identify elevated concentrations for the purposes of intervention. Surface measurements tell us what pesticide residues are present in an environment and at what concentrations. With appropriate transfer coefficients and activity data, these measurements can be used to estimate dermal and nondietary ingestion exposure.

Although exposure potential is highest during the first few days following an application, pesticide residues introduced into the indoor residential environment may persist for months or even years on surfaces or embedded in carpets, where these are protected from sunlight, rain, temperature extremes, and microbial action (Lewis *et al.,* 1994). Surface residues may contribute to the exposure of household occupants through multiple routes: dermal absorption, inhalation of resuspended particles, nondietary ingestion of residues adhering to mouthed objects and skin, and dietary ingestion resulting from children's unique handling of food (Butte and Heinzow, 2002). Oral ingestion and dermal absorption of surface residues may be major routes of exposure for infants and toddlers who spend much of their time on the floor, explore their world through mouthing, experience frequent hand-to-mouth and object-to-mouth contacts, and who may have pica tendencies (Butte and Heinzow, 2002; Cohen Hubal *et al.,* 2000a, b; Freeman *et al.,* 2004; Lewis *et al.,* 1994; Tulve *et al.,* 2002). Ingestion of soil is also a special concern for young children, who may ingest up to 10 times more soil than adults on a per kilogram body weight basis (LaGoy, 1987).

Several surface sampling methods exist including deposition coupons, Octadecyl (C18) surface press sampler (EL Sampler), Lioy-Weisel-Wainman (LWW) sampler, vacuum, drag bar, California-roller, PUF roller, and surface wipes. These methods are generally classified by the degree to which they remove residues from surfaces: total available residue, transferable residue, and dust (Lewis, 2001). T*otal available residue* methods attempt to measure the total amount of contaminant on a surface (often with the aid of isopropanol as a solvent), *transferable residue* methods are intended to represent the amount that is transferred as a result of contact with the contaminated surface, and *dust collection* methods use a vacuum to collect dust-borne residue on surfaces and from carpet. Transferable residues are also referred to as dislodgeable residues. All studies discussed in this chapter employed more than one sampling method for surface measurements. Table 4.1 lists the studies that collected surface measurements along with the type of measurement taken. Limits of detection for each chemical by study and method are listed in Table 4.2.

Several variables may influence measured dust concentrations or surface loadings of pesticide residues. These variables include the collection method itself, surface type, compound physico-chemical characteristics, application method, application frequency, sampling locations, participant activities, and analytical capabilities. This chapter examines how these factors may have affected the surface residue measurements in the children's exposure measurement program, the implications for interpreting the data, and the consequences for exposure estimates.

Table 4.1 Studies and sample collection methods for surface measurements.

Study	Dust (ng/g)	Dust Load (ng/cm^2)	Soil (ng/g)	Total Surface Load (ng/cm^2)	Transferable Residues (ng/cm^2)
NHEXAS-AZ	✓	✓	✓	--	Wipes (water)
MNCPES				LWW	C18 Press
CTEPP	✓	✓	✓	--	Wipes (2 mL IPA), PUF Roller
CCC	--	--	✓	Wipes (20 mL IPA)	C18 Press
JAX	--	--	--	Wipes (20 mL IPA)	C18 Press
CHAMACOS	✓	✓	✓	Wipes (20 mL IPA)	C18 Press
CPPAES	--	--	--	Deposition Coupons, LWW	--
Test House	--	--	--	Deposition Coupons, Wipes (10 mL IPA)	PUF Roller C18 Press
PET	✓	--	✓		PUF Roller
DIYC	--	--	--	Wipes (20 mL IPA)	PUF Roller
Daycare	--	--	--	Wipes (20 mL IPA)	PUF Roller, C18 Press

--, matrix not sampled
LWW, Lioy-Weisel-Wainman sampler
C18, 3M Empore™ Octadecyl (C18) filters
PUF, Polyurethane foam

Table 4.2 Limits of detection (ng/g or ng/cm²) for surface measurements by study, method, and compound.

Study	Method	Chlor-pyrifos	Diaz-inon	*c*-Per-methrin	*t*-Per-methrin	Cyflu-thrin	Cyper-methrin	Esfen-valerate	TCPy	IMP
Soil (ng/g)										
MNCPES	Soil	10	10	10	10	--	--	--	--	--
CTEPP	Soil	0.5	0.5	0.5	0.5	5	--	--	0.2	0.2
CCC	Soil	5	2	5	5	6	6	--	--	--
PET	Soil	--	60	--	--	--	--	--	--	--
Dust (ng/cm² or ng/g)										
NHEXAS-AZ	Dust (ng/cm²)	0.002	0.002	--	--	--	--	--	--	--
CTEPP	Dust (ng/cm²)	0.0003	0.0003	0.0003	0.0003	0.0030	--	--	0.0003	--
NHEXAS-AZ	Dust (ng/g)	4	18	--	--	--	--	--	--	--
CTEPP	Dust (ng/g)	2	2	2	2	10	--	--	2	2
CHAMACOS	Dust (ng/g)	1	1	1	1	100	--	--	--	--
PET	Dust (ng/g)	--	60	--	--	--	--	--	--	--
Total Available Residue (ng/cm²)										
NHEXAS-AZ	IPA Wipe	0.070	2.00	--	--	--	--	--	--	--
MNCPES	LWW	1.200	3.50	--	--	--	--	--	--	--
CCC	IPA Wipe	0.005	0.002	0.005	0.005	0.006	0.006	--	--	--
JAX	IPA Wipe	0.005	0.002	0.005	0.005	0.006	0.006	0.008	--	--
CHAMACOS	IPA Wipe	0.005	0.005	0.005	0. 002	--	--	--	--	--
CPPAES	IPA Wipe	0.001	--	--	--	--	--	--	--	--
CPPAES	LWW	0.030	--	--	--	--	--	--	--	--
CPPAES	Dep Coup	0.010	--	--	--	--	--	--	--	--
TESTHOUSE	IPA Wipe	0.001	--	--	--	--	--	--	--	--
TESTHOUSE	Dep Coup	0.010	--	--	--	--	--	--	--	--
DIYC	IPA Wipe	--	0.300	--	--	--	--	--	--	--
DAYCARE	IPA Wipe	--	--	--	--	--	--	0.400	--	--
Transferable Residue (ng/cm²)										
MNCPES	C18 Press	0.330	0.140	--	--	--	--	--	--	--
CTEPP	IPA Wipe	0.0007	0.0007	0.0007	0.0007	0.007	--	--	0.0007	0.0007
CTEPP	PUF	0.0004	0.0004	0.0004	0.0004	0.004	--	--	0.0004	0.0004
TESTHOUSE	C18 Press	0.030	--	--	--	--	--	--	--	--
TESTHOUSE	PUF	0.001	--	--	--	--	--	--	--	--
PET	PUF	--	0.030	--	--	--	--	--	--	--
DIYC	C18 Press	--	1.200	--	--	--	--	--	--	--

--, analyte not measured

4.2 Dust and Soil Measurements

Dust is considered a repository of environmental pollutants that have accumulated indoors from both internal and external sources. Dust collected by vacuum is usually sieved to retain a particular size fraction for analysis, which may have important implications since pesticide concentrations are inversely related to particle size (Lewis *et al.,* 1999). Measurements in dust may be reported as concentrations (mass residue per unit weight of dust, ng/g) or as loadings (mass residue per unit area sampled, ng/cm^2). There is a lack of consensus on which of these metrics is more relevant to human exposure to pesticides; however, lead studies have suggested that lead loading correlates better with children's blood lead levels than does lead concentration (Lanphear, 1995).

Pesticides were measured in dust samples from the NHEXAS-AZ, CTEPP, CHAMACOS and PET studies. The CTEPP, CHAMACOS, and PET studies used the High Volume Small Surface Sampler (HVS3), whereas NHEXAS-AZ used a modified commercially available vacuum for ease of sample collection. The HVS3 was developed for the EPA and efficiently collects carpet-embedded dust retaining the associated pesticides (Roberts *et al.,* 1991; Lewis *et al.,* 1994). The HVS3 is a high-powered vacuum cleaner equipped with a nozzle that can be adjusted to a specific static pressure and air flow rate. A cyclone removes particles >5 μm from the air stream for collection in a catch bottle. Use of this sampler is limited to floors or other large flat surfaces (Roberts *et al.,* 1991; Ness, 1994; Lewis *et al.,* 1994). The ASTM (American Society for Testing and Materials) method for the collection of carpet-embedded dust requires an apparatus with the specifications of the HVS3 (ASTM, 1993). Pesticide concentrations in soil were measured in the same studies and results have been included in this chapter to allow comparisons between indoor and outdoor exposure pathways for the same children.

Pesticide Presence in Dust and Soil

Detection limits are listed in Table 4.2. Detection frequencies are presented in Figure 4.1 for soil samples and Figure 4.2 for dust samples. Concentrations of pesticides in soil and dust samples at the median and 95th percentile are listed in Table 4.3 (complete summary statistics are listed in Tables A.8 through A.19 in Appendix A).

- With the exception of cyfluthrin (for which analytical difficulties produced a higher detection limit), dust samples had high detection frequencies (>95%) in CTEPP and CHAMACOS. Detection frequencies were lower in NHEXAS-AZ due to higher detection limits.
- The high detection frequencies of pesticides observed in dust across studies is consistent with dust being a repository of contaminants.
- Detection frequencies for soil samples, on the other hand, were generally low (Figure 4.1). The high detection frequency of diazinon in PET study soil was due to direct lawn applications of the pesticide prior to sample collection.
- Pesticide concentrations were much lower in soil samples than in dust samples. In general, soil levels at the 95th percentile were a factor of 10 to 100 times lower than dust levels at the same percentile. This result suggests that in the absence of outdoor turf treatments, ingestion of soil may not be an important exposure pathway for these

pesticides, with the possible exception of children exhibiting pica behavior.

Concentrations in Dust and Soil: Summary Findings

Lognormal probability plots that graphically depict pesticide concentrations in soil from large observational field studies are presented in Figure 4.3. Plots that depict pesticide concentrations and loadings in dust are given in Figures 4.4 and 4.5. Box-and-whisker plots comparing pesticide concentrations and loadings in dust across all studies are given in Figures 4.6 and 4.7.

- The upper tails of the soil concentration distributions tend to be in the same range as the lower tails of the dust concentration distributions (Figures 4.3-4.5). For example, the 95th percentile for both chlorpyrifos and diazinon in *soil* is approximately 10 ng/g, and the 5th percentile for both of these compounds in *dust* is also near 10 ng/g.
- Among the pesticides measured in soil, cyfluthrin stands out for its high values at the 95th percentile (Table 4.3). Due to the low detection frequencies, no additional analysis was conducted with the soil data.
- Comparisons of concentrations in dust across studies (Figures 4.4-4.5) show permethrin (a pyrethroid) to be about an order of magnitude higher than chlorpyrifos and diazinon (both organophosphates).
- Overall, diazinon concentrations are lower than all other pesticides reported in dust, as illustrated in the box-and-whisker plots (Figures 4.6-4.7).
- High loadings of diazinon in indoor house dust following the lawn treatment in the PET study suggest translocation into the house by the occupants and their pets.
- The concentration ranking among the compounds in dust is the opposite of that found in air where the more volatile pesticides showed the higher concentrations. The less volatile pyrethroid pesticides tend to partition to the dust and may degrade more slowly, allowing accumulation over time from repeated applications. These results point to the importance of dust as a primary residential exposure medium for the less volatile pesticides. In addition, the exposure factors that are important for other nonvolatile contaminants such as lead (Melnyk *et al.*, 2000) may also be important for the less volatile pesticides.
- In general, the lognormal plots (Figures 4.4-4.5) indicate that differences between study populations are more apparent with dust loadings than with dust concentrations.
- In CTEPP, pesticide loadings in surface dust (ng/cm^2) were higher in daycare centers (DC) than in homes (HM) (Figures 4.6-4.7). This appears to be a function of the amount of surface dust present, as the pesticide concentrations in the dust do not differ by much (Figures 4.6-4.7). Studies with lead have suggested that loading has a greater impact than concentration on intake, and the same may or may not be true for pesticides.
- Concentrations of chlorpyrifos in dust (ng/g) are similar across studies (Figure 4.4) suggesting that the usage of chlorpyrifos did not change significantly from the timeframe of the NHEXAS-AZ study (1995-1997) to the CTEPP study (2000-2001).
- As with the other surface measurement methods, *cis*- and *trans*-permethrin have similar concentration profiles in dust samples.

Table 4.3 Median and 95th percentile values for soil (ng/g) and dust (ng/cm² and ng/g) measurements by study.

	Units	Chlorpyrifos		Diazinon		*c*-Permethrin		*t*-Permethrin		Cyfluthrin		TCPy		IMP	
		P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
SOIL															
MNCPES	ng/g	<10.0	<10.0	<10.0	<10.0	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h[a]	ng/g	<0.5	17.0	<0.5	4.2	<0.5	13.0	<0.5	18.0	<5.0	32.0	0.6	11.0	--	--
CTEPP-NC d	ng/g	<0.5	0.8	<0.5	<0.5	<0.5	2.6	<0.5	2.2	<5.0	42.0	<0.2	1.2	--	--
CTEPP-OH h	ng/g	<0.5	14.0	<0.5	4.7	<0.5	2.7	<0.5	2.1	<5.0	64.0	0.7	8.9	<0.2	2.1
CTEPP-OH d	ng/g	<0.5	6.2	<0.5	7.1	<0.5	<0.5	<0.5	<0.5	<5.0	42.0	0.6	6.3	<0.2	1.4
CCC	ng/g	<5.0	27.0	<2.0	22.0	<5.0	8.6	<5.0	12	<6.0	8.6	--	--	--	--
PET	ng/g	--	--	22000	50000	--	--	--	--	--	--	--	--	--	--
DUST (Loadings)															
NHEXAS-AZ	ng/cm²	0.007	2.80	0.002	0.18	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h	ng/cm²	0.009	0.42	0.002	0.12	0.10	4.90	0.09	4.40	<0.003	0.16	0.008	0.37	--	--
CTEPP-NC d	ng/cm²	0.066	1.30	0.026	9.90	0.69	5.50	0.41	6.30	<0.003	0.60	0.020	0.37	--	--
CTEPP-OH h	ng/cm²	0.006	0.35	0.002	0.31	0.05	3.80	0.03	3.90	0.018	0.25	0.004	0.16	0.001	0.046
CTEPP-OH d	ng/cm²	0.046	0.89	0.022	0.39	0.27	4.80	0.31	4.70	0.140	1.10	0.024	0.40	0.004	0.072
PET	ng/cm²	--	--	0.350	68	--	--	--	--	--	--	--	--	--	--
DUST (Concentrations)															
NHEXAS-AZ	ng/g	140	120000	150	8000	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h	ng/g	130	1200	18	390	800	21000	630	19000	47	1700	96	1100	--	--
CTEPP-NC d	ng/g	140	920	47	6900	890	10400	760	12000	79	1500	63	300	--	--
CTEPP-OH h	ng/g	52	1400	20	1700	470	7600	340	9200	200	1300	41	820	14	
CTEPP-OH d	ng/g	180	1100	38	1600	690	3800	480	3400	350	890	67	500	17	310
CHAMACOS	ng/g	49	1200	21	820	150	2900	40	15000	<50	303.6	--	--	--	--
PET	ng/g	--	--	3100	150000	--	--	--	--	--	--	--	--	--	--

[a] CTEPP: h = home, d = daycare

--, analyte not measured

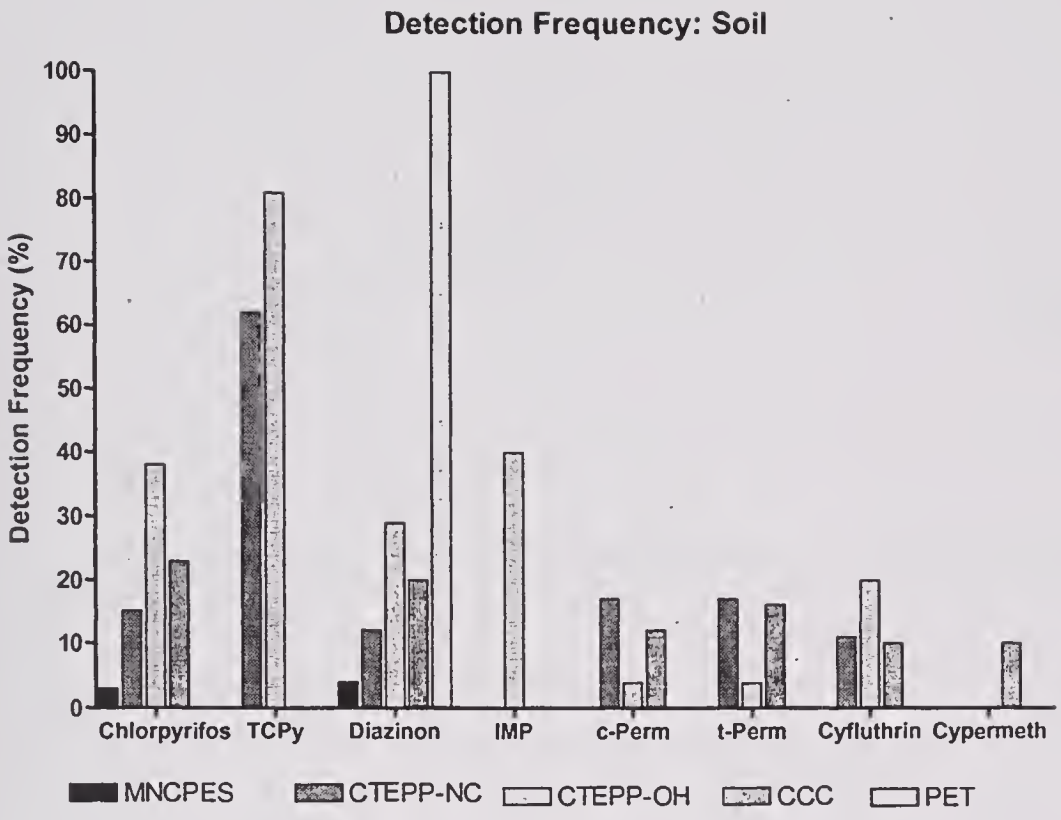


Figure 4.1 Detection frequencies of pesticides and degradates in soil.

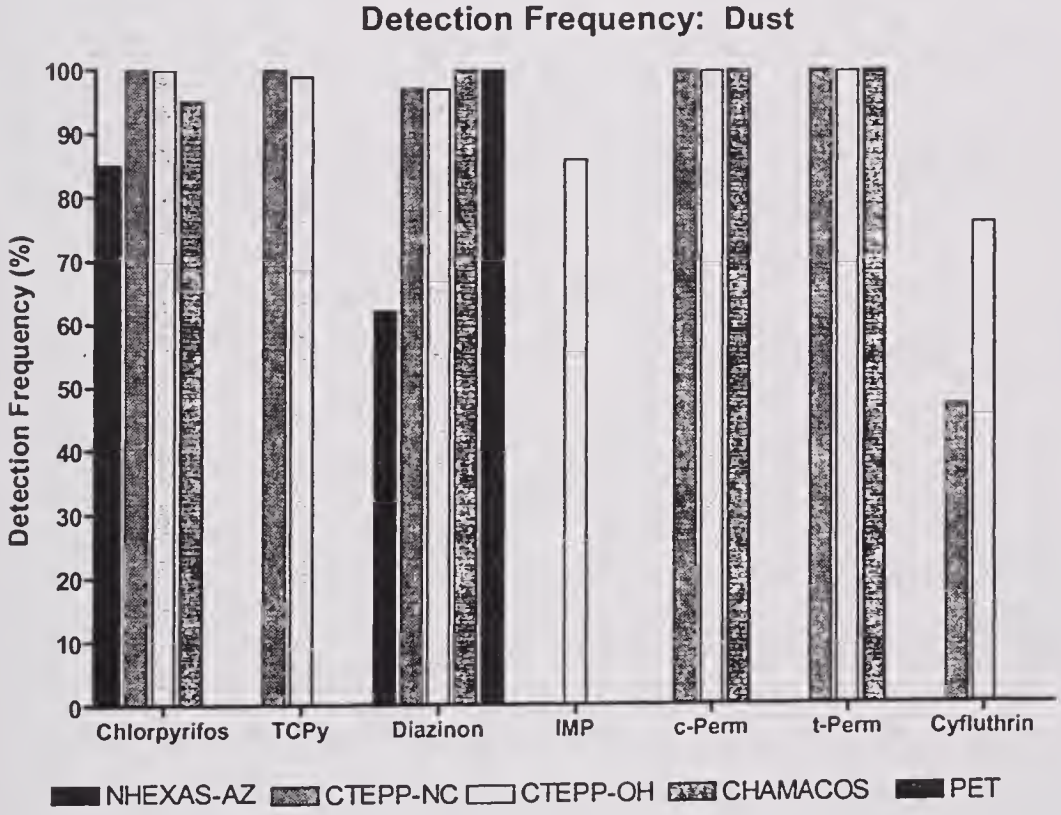


Figure 4.2 Detection frequencies of pesticides and degradates in dust.

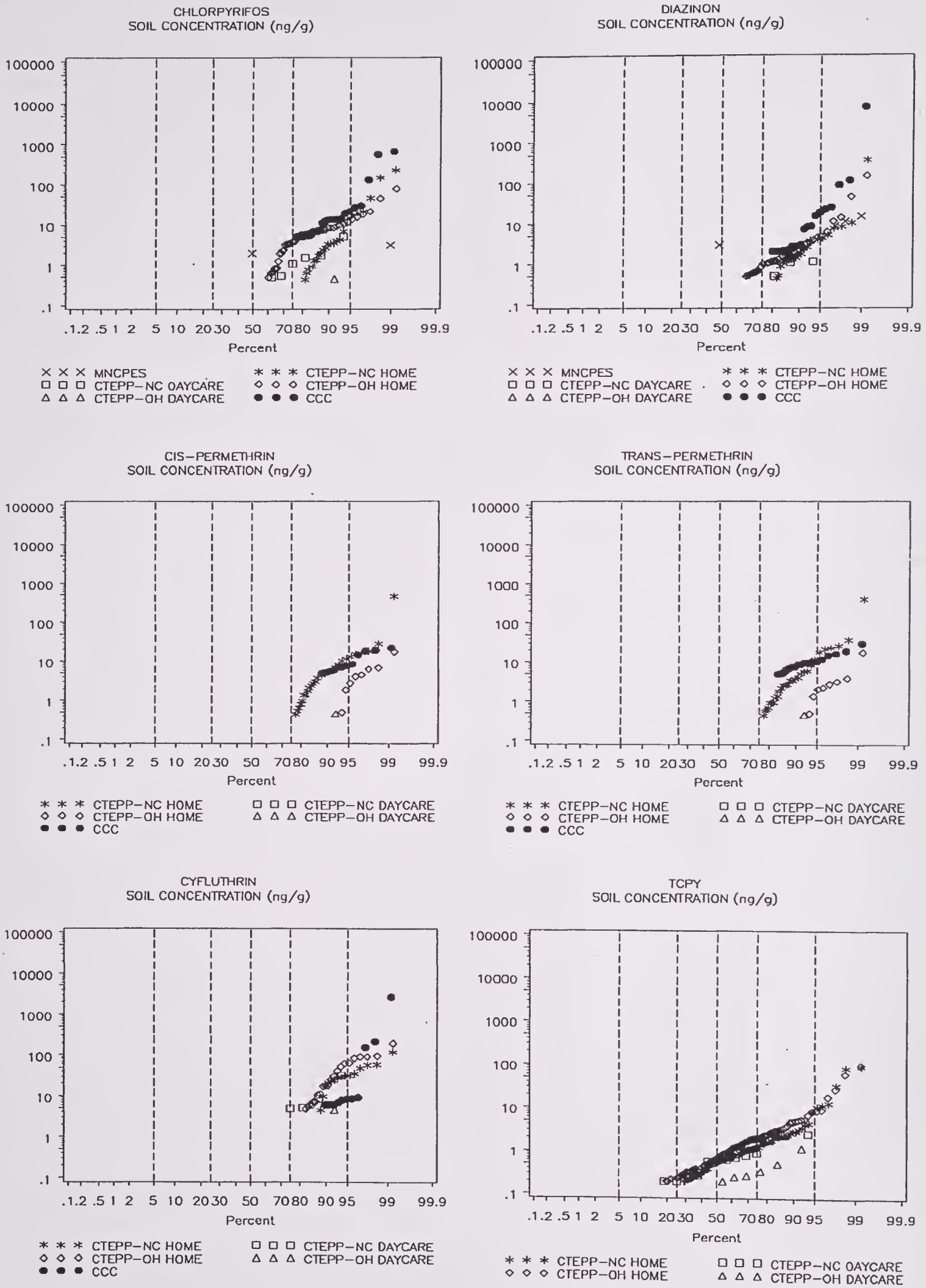
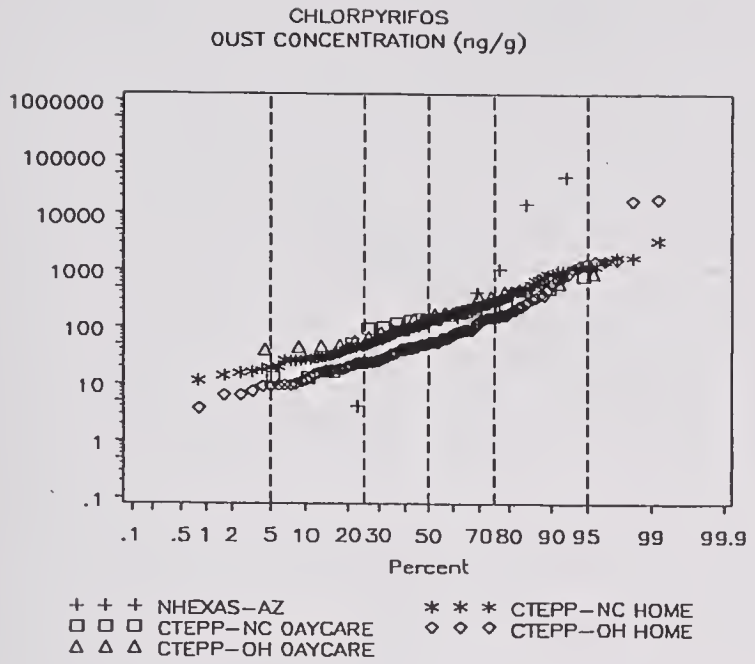
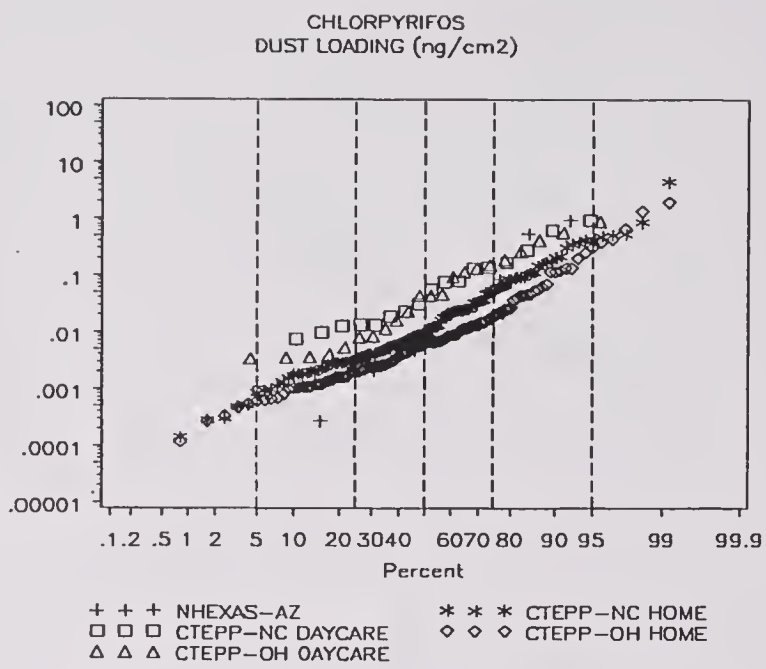
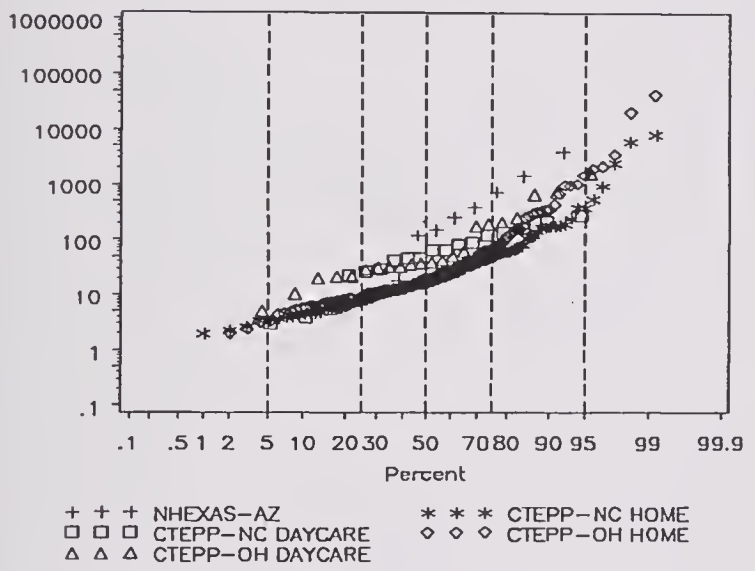
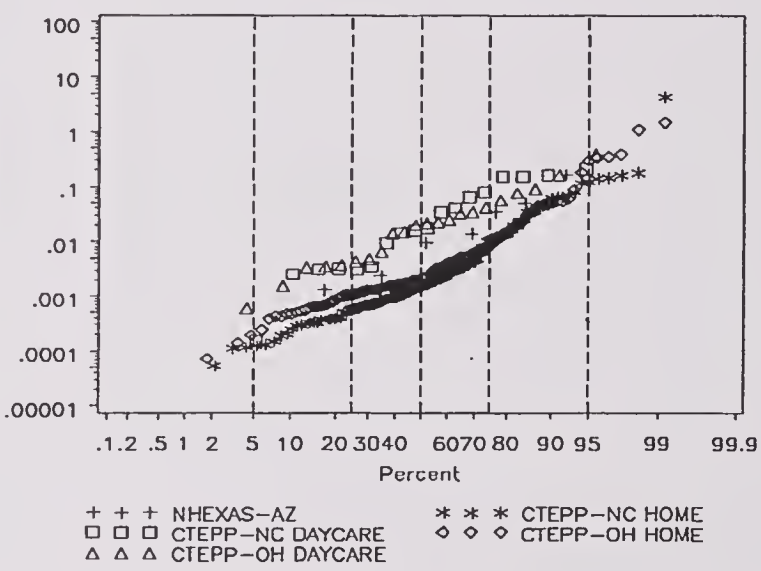


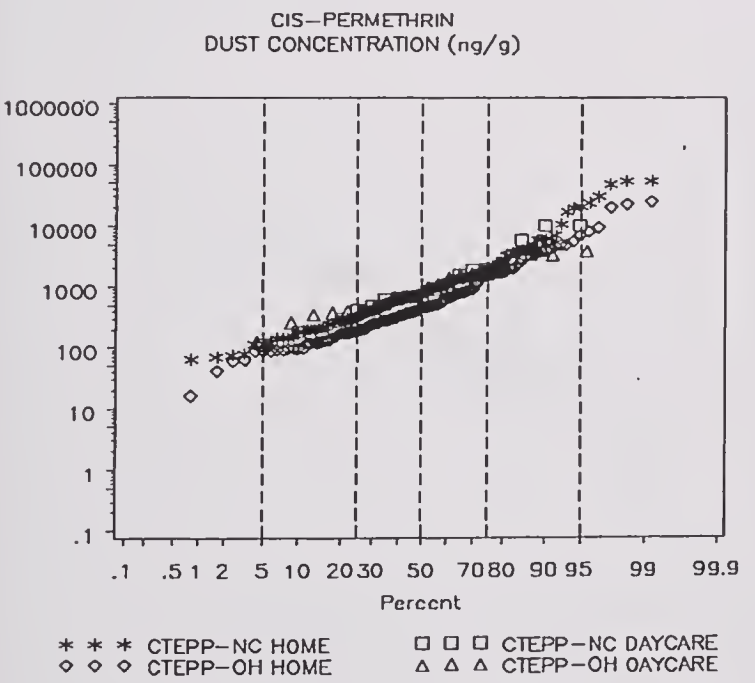
Figure 4.3 Lognormal probability plots of soil concentrations (ng/g) for chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cyfluthrin, and TCPy.











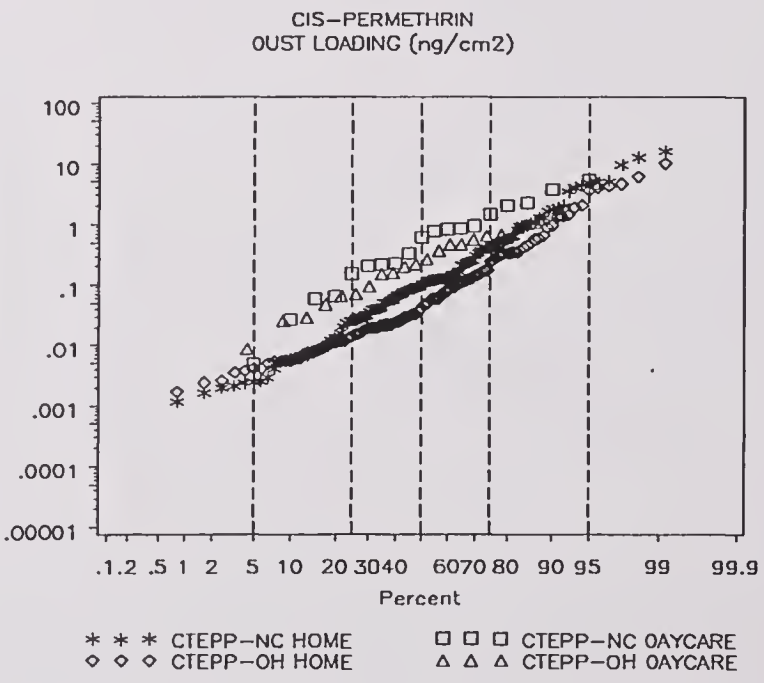


Figure 4.4 Lognormal probability plots of dust concentrations (ng/g) and loadings (ng/cm²) for chlorpyrifos, diazinon, and *cis*-permethrin.

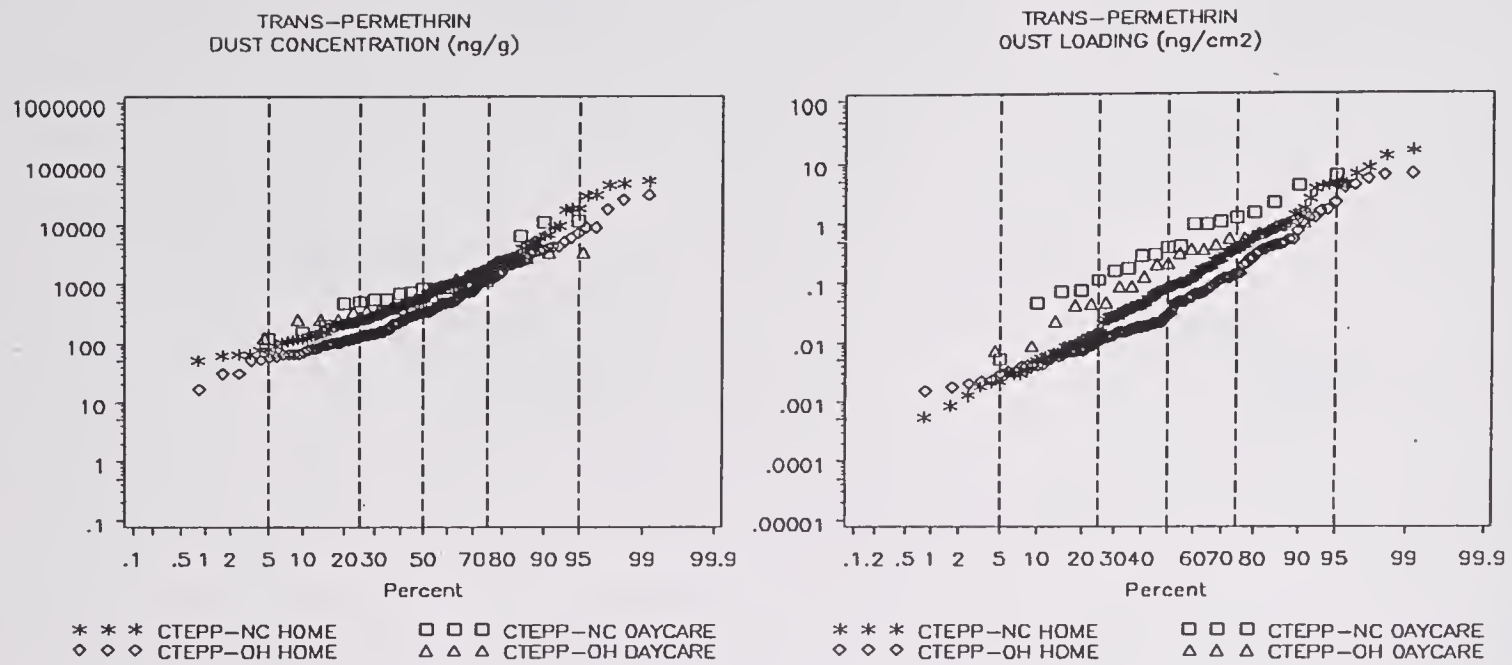


Figure 4.5 Lognormal probability plots of dust concentrations (ng/g) and loadings (ng/cm²) for *trans*-permethrin, cyfluthrin, and TCPy.

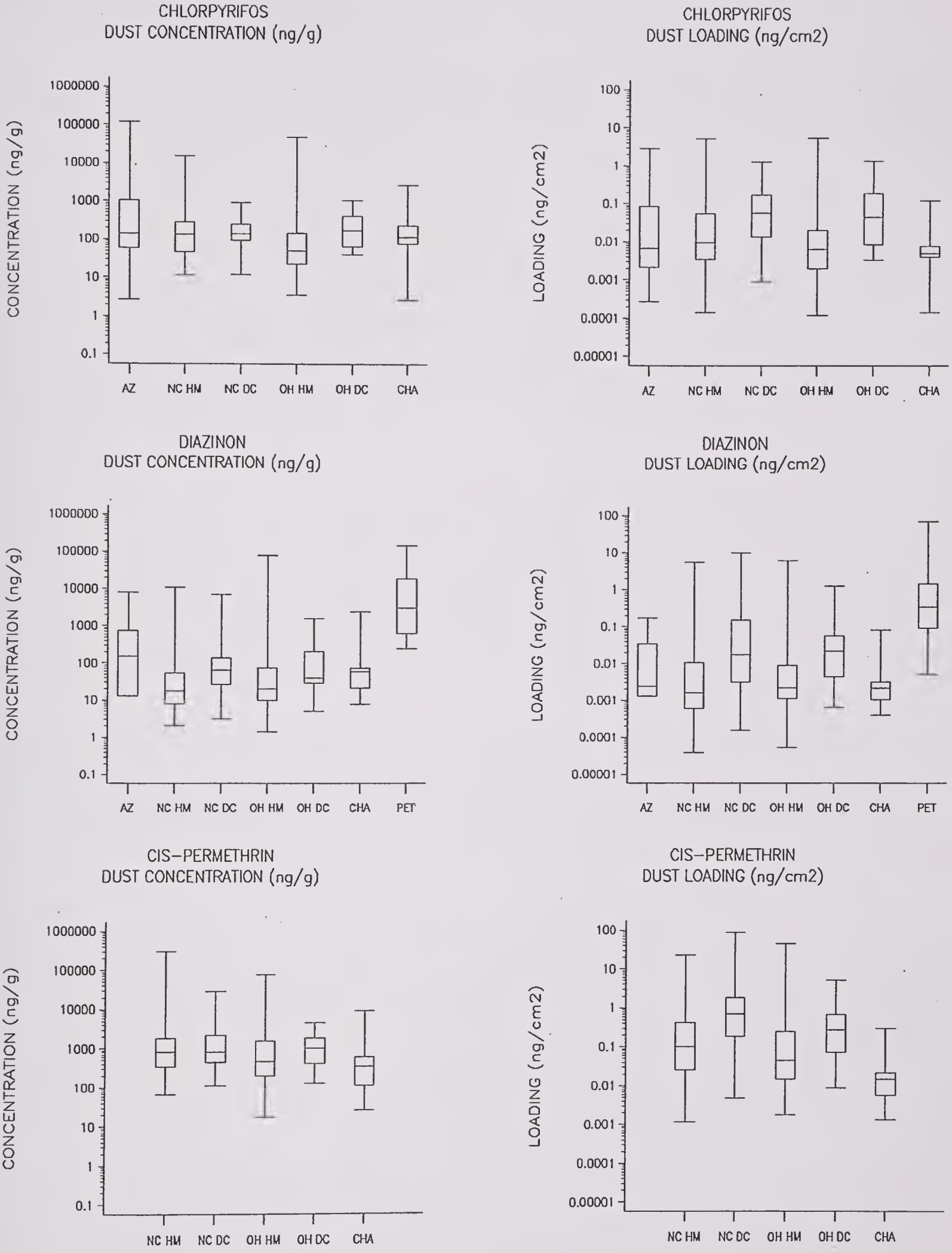


Figure 4.6 Box-and-whisker plots of dust concentrations (ng/g) and loadings (ng/cm²) for chlorpyrifos, diazinon, and *cis*-permethrin.

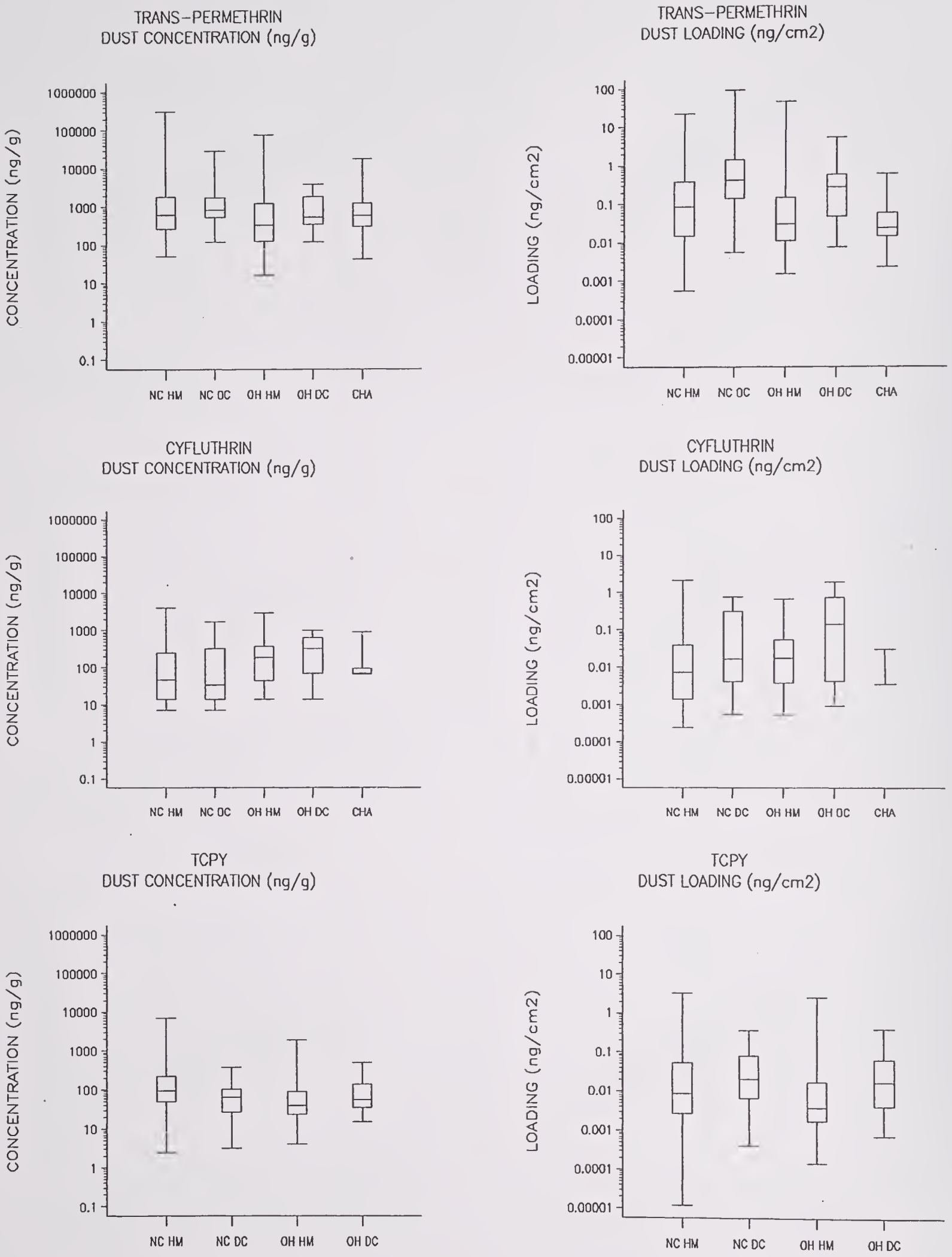


Figure 4.7 Box-and-whisker plots of dust concentrations (ng/g) and loadings (ng/cm²) for *trans*-permethrin, cyfluthrin, and TCPy.

4.3 Total Available Residue Measurements

Total available residue methods are intended to measure the total amount of contaminant on a surface. These methods involve either a solvent-assisted mechanical (wiping) action or the stationary capture of descending airborne droplets and particles. Total available residue loadings were measured in:

- NHEXAS-AZ using the LWW sampler,
- MNCPES using the LWW sampler,
- CCC from the floors and other surfaces (*e.g.*, counters, desktops) using surface wipes,
- JAX from the floor in the application area using surface wipes,
- CHAMOCOS using surface wipes,
- CPPAES using the LWW and deposition coupons,
- Test House using deposition coupons and surface wipes,
- DIYC using surface wipes, and
- Daycare using surface wipes.

The Lioy-Weisel-Wainman (LWW) sampler (Patent #RWJ-91-28) was developed to quantitatively measure dust on smooth surfaces and has been validated in laboratory and field tests (Lioy *et al.,* 1993; Freeman *et al.,* 1996). The LWW sampler achieves quantitative wipe collection using a movable constant pressure block within a template marking a specific area of 100 cm^2. Octadecyl-bonded (C18) disks that have been immersed in isopropyl alcohol are attached to a silicon rubber pad on the block. More details about this sampler can be found in Gurunathan *et al.* (1998) and Hore (2003).

Surface wipes are typically surgical dressing sponges wetted with isopropyl alcohol (IPA). The sponge is wiped multi-directionally through a defined area in an S-shaped configuration. Floor locations where young children may spend the most amount of time are usually selected. Residue loadings on irregularly shaped objects such as toys that are frequently handled by children (for estimating indirect ingestion exposures) are also measured using the wipe method.

Deposition coupons are used to estimate surface loadings of airborne and dust-bound residues that "settle out" of the air following an application (Ness, 1994). These consist of a sorptive material (*e.g.*, cotton, sponge, rayon) with a non-sorptive backing (aluminum foil) (Stout and Mason, 2003) and are placed in locations where the coupons will not be disturbed. Coupons may be repeatedly collected and replaced (interval) or collected only at the end of the sampling event (cumulative). Both interval and cumulative types were collected in CPPAES, whereas only interval deposition coupons were used in the Test House.

Pesticide Presence in Total Available Residues

Limits of detection for each chemical by study are given above in Table 4.2. Detection frequencies are given in Figure 4.8.

- The limits of detection varied widely among studies, but are similar within a study for both organophosphate and pyrethroid pesticides.
- Following dust methods, total available residue methods have the lowest limits for detection.
- Detection frequencies were slightly higher for the organophosphate pesticides in two of the three studies where both OP and pyrethroid pesticides were measured.
- Detection frequencies were higher in the smaller, focused studies than in the survey studies due to timing of the measurements with respect to recent applications.

Total Available Residues: Summary Findings

Surface loadings for the median and 95th percentile are listed in Table 4.4 for all of the pesticides that were detected across studies (complete summary statistics are listed in Tables A.20 through A.24 in Appendix A). Lognormal probability plots are presented in Figure 4.9 for the most frequently detected pesticides which include chlorpyrifos, diazinon, *cis*- and *trans*-permethrin, cyfluthrin, and cypermethrin. The MNCPES data are not included because of the comparatively high detection limit and low detection frequencies. Box and whisker plots that graphically depict the total available residue loading results from all studies are given in Figure 4.10.

- In wipe samples, permethrin levels reported at the 95th percentile were approximately an order of magnitude higher than chlorpyrifos and diazinon levels at the 95th percentile (Table 4.4).
- Levels of diazinon and esfenvalerate reported at the 95th percentile were at least an order of magnitude higher in studies with a known application (DIYC, Daycare) than in the survey studies (CCC, JAX-Screening).
- The lognormal probability plots (Figure 4.9) show that loadings of all frequently detected pesticides are substantially higher in the JAX screening wipe samples than in the CCC and CHAMACOS wipe samples.
- The total available residue distributions (Figure 4.9) of chlorpyrifos and *cis*- and *trans*-permethrin are relatively similar to each other within a specific large observational field study.
- Cypermethrin loadings tend to be the highest and diazinon loadings tend to be the lowest (Figure 4.9) of the pesticides of interest in the large observational field studies.
- The boxplots (Figure 4.10) reveal that chlorpyrifos, diazinon, and esfenvalerate loadings are substantially higher in those studies with a known application (CPPAES, Test House, DIYC, and Daycare).

- Low cyfluthrin loadings in wipe samples in Figure 4.9 (substantially lower than all other pesticide residues) suggest that cyfluthrin may not have been routinely used for pest treatment.
- MNCPES and CPPAES are the only studies that employed the LWW. The chlorpyrifos loadings measured in CPPAES were significantly higher (ANOVA, $p=0.002$, test results not presented) due to known pesticide applications coinciding with the sampling period.
- Although the MNCPES measurements did not coincide with a pesticide application, 62% of the LWW samples had detectable levels of chlorpyrifos, suggesting that chlorpyrifos remains on residential surfaces for a long period of time. It is unclear, however, how much of this is readily available for transfer and how much is freed from the pores and/or body material of the surfaces by the mechanical and solvent action of the LWW sampler.
- Mean post-application deposition coupon levels were significantly higher in the Test House than in CPPAES (ANOVA, $p<0.0001$, test results not presented). Factors responsible may include the following: three CPPAES homes received applications with only trace chlorpyrifos concentrations; the application performed in the Test House may have been more thorough than applications in the CPPAES homes; the Test House may have had a higher application of active ingredient per effective volume of the home (see Section 3.6), and some of the CPPAES occupants reported cleaning their homes and/or intentionally increasing ventilation after application, thereby reducing the amount of chlorpyrifos available for movement and capture on a deposition coupon.
- In studies (*e.g.*, CPPAES) where surface wipe samples were collected both pre- and post-application of a semi-volatile pesticide such as chlorpyrifos, the post-application pesticide loadings were higher than the pre-application values, including on surfaces that did not receive a direct application. This suggests that semi-volatile pesticides rapidly translocate from application surfaces to adjacent surfaces. We do not yet have information on the speed or extent of translocation for less volatile pesticides like pyrethroids.
- Two types of locations were sampled in JAX, the application area and a play area. In general, the surface residue loadings were higher at the application area than at the play area.
- The surface wipe samples collected in the CCC study were collected from two locations in each of the randomly selected rooms of the child care centers: a floor and desk top/table top surface. In general, the floor residue loadings were higher.

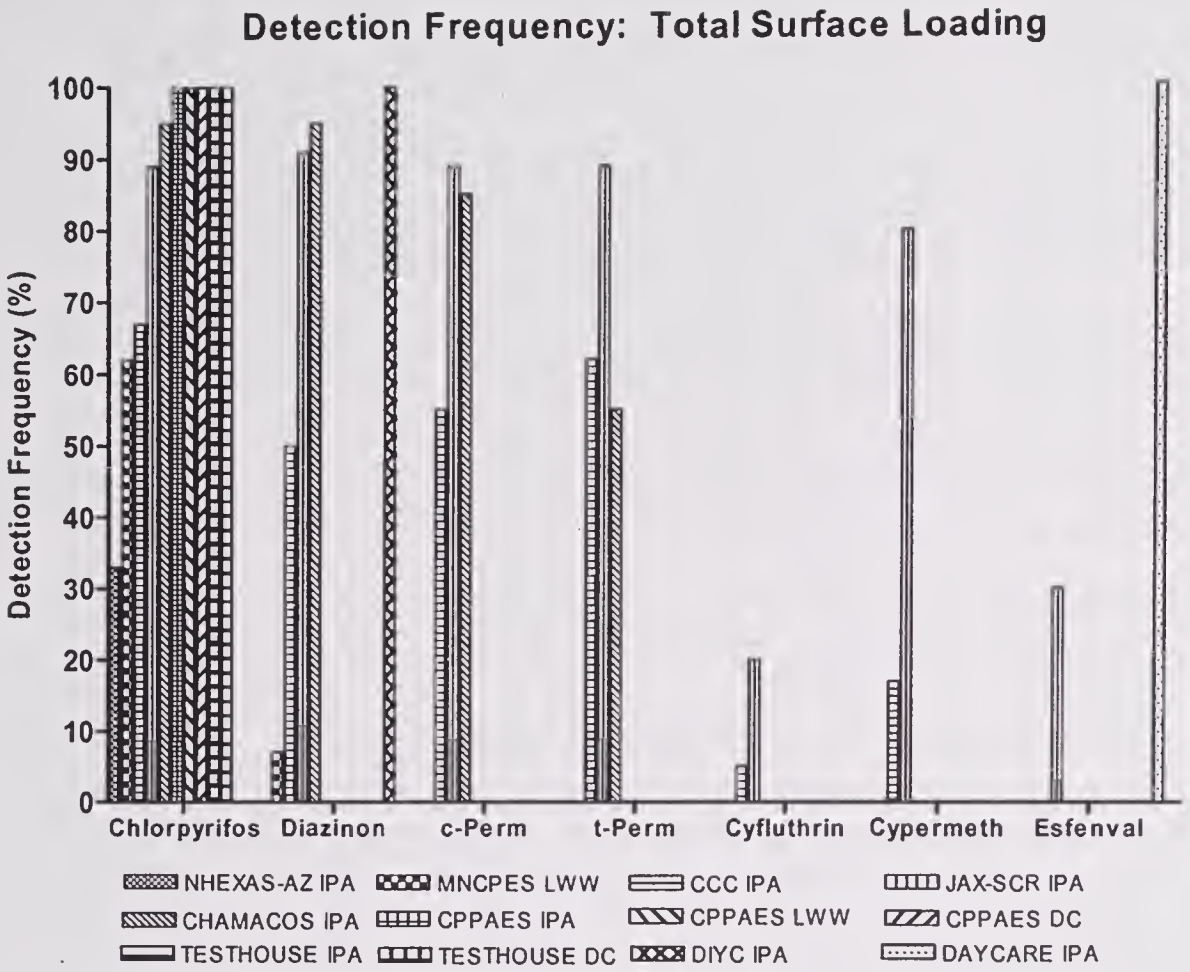


Figure 4.8 Detection frequencies for pesticides using total available residue collection methods.

Table 4.4 Median and 95th percentile values for total available residues (ng/cm²) by study.

Study	Method	Chlorpyrifos		Diazinon		*c*-Permethrin		*t*-Permethrin		Cyfluthrin		Cypermethrin		Esfenvalerate	
		P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
NHEXAS-AZ	IPA Wipe	<0.07	7.5	<2.000	<2.0	--	--	--	--	--	--	--	--	--	--
MNCPES	LWW	1.20	1.5	<3.500	3.5	--	--	--	--	--	--	--	--	--	--
CCC	IPA Wipe	0.03	0.9	0.002	0.5	0.009	0.67	0.02	1.1	<0.006	0.08	<0.006	0.8		
JAX-SCR	IPA Wipe	0.53	10.0	0.110	3.3	2.200	32.00	2.90	40.0	<0.006	4.30	2.600	750.0	<0.008	3.5
JAX-AGG	IPA Wipe	0.10	3.1	<0.002	4.0	0.210	42.00	0.26	67.0	<0.006	10.00	--	--	--	--
CHAMACOS	IPA Wipe	0.05	0.2	0.040	0.1	0.100	1.70	0.20	3.6	<0.050	0.40	--	--	--	--
CPPAES Pre	LWW	0.17	1.3	--	--	--	--	--	--	--	--	--	--	--	--
CPPAES	LWW	0.61	10.0	--	--	--	--	--	--	--	--	--	--	--	--
CPPAES	IPA Wipe	0.03	0.2	--	--	--	--	--	--	--	--	--	--	--	--
CPPAES	Dep Coup	1.40	9.6	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE Pre	IPA Wipe	4.70	9.1	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE	IPA Wipe	11.00	36.0	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE	Dep Coup	3.20	62.0	--	--	--	--	--	--	--	--	--	--	--	--
DIYC Pre	IPA Wipe	--	--	3.8	21.0	--	--	--	--	--	--	--	--	--	--
DIYC	IPA Wipe	--	--	5.5	72.0	--	--	--	--	--	--	--	--	--	--
DAYCARE	IPA Wipe	--	--	--	--	--	--	--	--	--	--	--	--	3.200	51.0

--, pesticide not measured

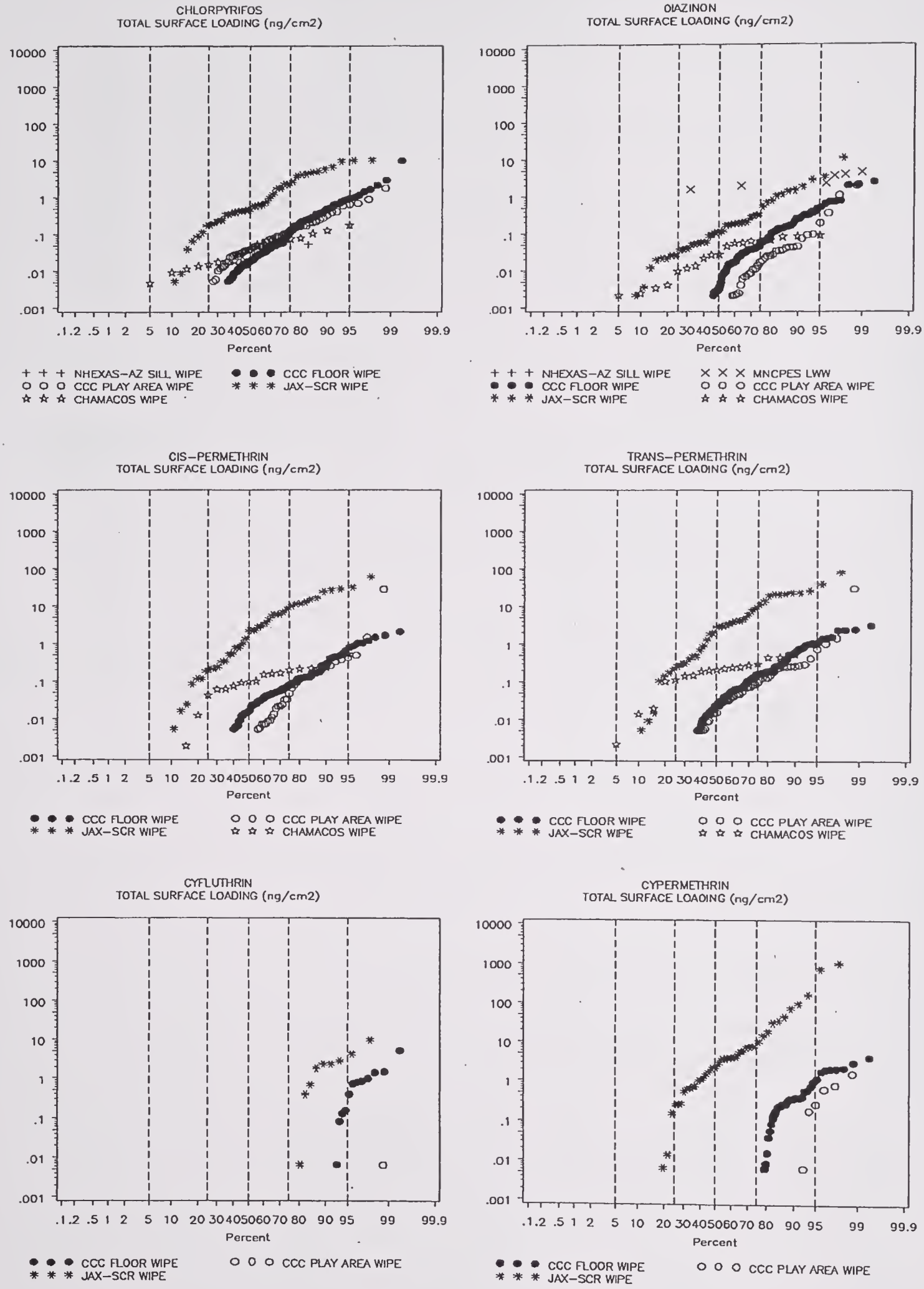


Figure 4.9 Lognormal probability plots for the most frequently detected pesticides which include chlorpyrifos, diazinon, *cis*- and *trans*-permethrin, cyfluthrin, and cypermethrin.

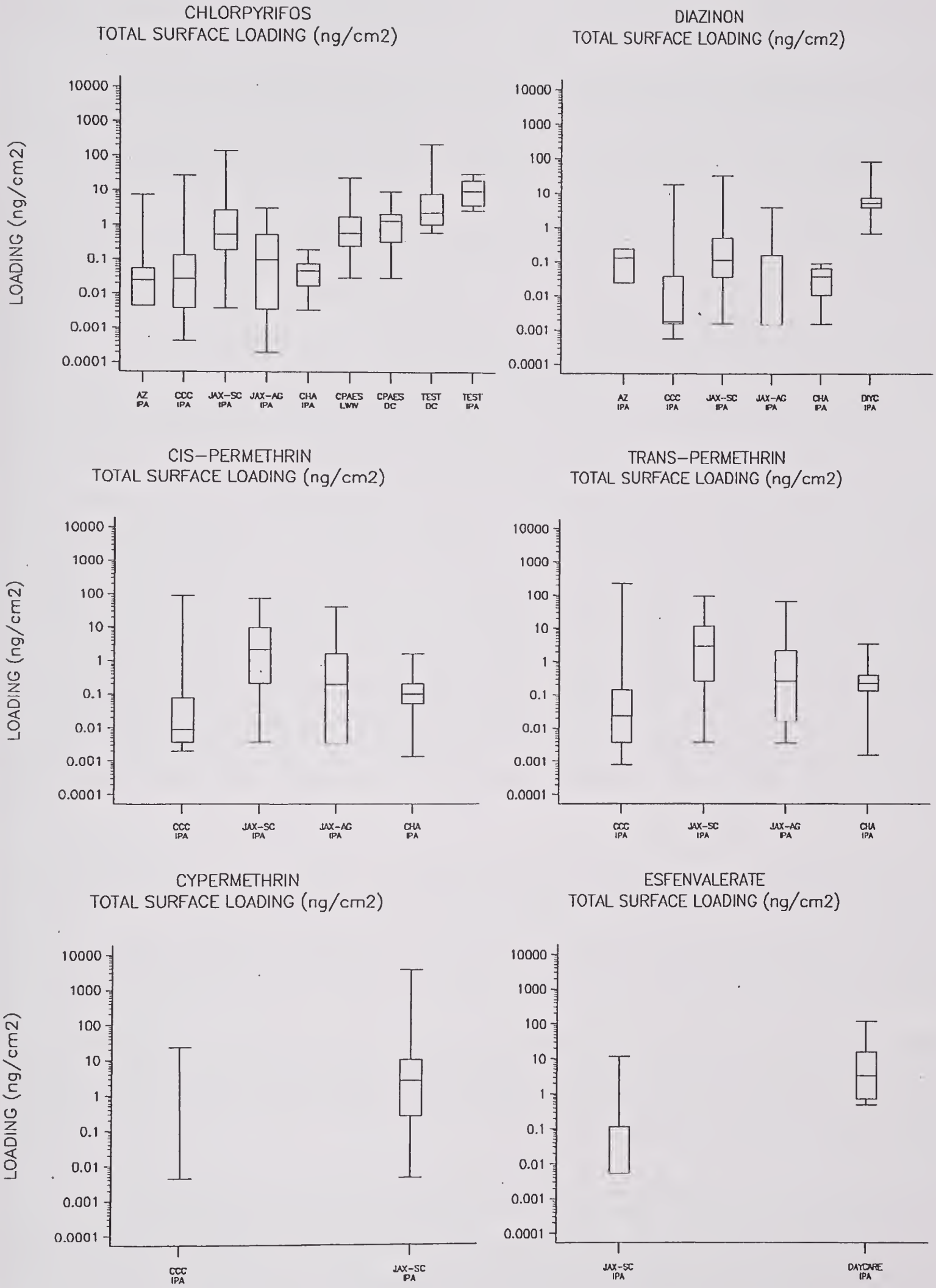


Figure 4.10 Box-and-whisker plots of total available residue surface loadings (ng/cm²) for chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cypermethrin, and esfenvalerate.

4.4 Transferable Residue Measurements

Transferable residue methods are intended to represent the surface loading that may be transferred as a result of contact with the contaminated surface; that is, instead of complete removal, they are typically intended to mimic transfer to skin during a single dermal contact with a surface, where transfer is aided by only saliva, sweat, or the sebum layer on the skin. Transferable residue loadings were measured in:

- MNCPES using the C18 press sampler on floors and non-floor surfaces,
- CTEPP using surface wipes with 2 mL 75% IPA on hard-surface floors and counters and a PUF roller on carpeted floors,
- CCC using the C18 press sampler on carpeted floors,
- JAX using the C18 press sampler on carpeted floors,
- CHAMACOS using the C18 press sampler on carpeted floors,
- Test House using the C18 press sampler and a PUF roller skin on carpeted floors,
- DIYC using the PUF roller on both hard-surface and carpeted floors, and
- Daycare using the C18 press sampler and the PUF roller on carpeted floors.

The Modified C18 Surface Press Sampler was based on the original EL Sampler designed by Edwards and Lioy to collect pesticides in house dust from carpeted floors (Edwards and Lioy, 1999; Hore, 2003). EPA modified the press sampler to use two 9-cm diameter sampling discs for a total sampling area of 114 cm^2 and eliminated the spring mechanism, henceforth it became known as the Modified C18 Surface Press Sampler. Unlike vacuum methods that collect household dust from all depths of the carpet pile and base, the surface press sampler is designed to only contact and remove residue from the surface. The developers maintain that the sampler replicates the collection efficiency of human skin and reflects transfer from single hand press (Edwards and Lioy, 1999; Lioy *et al.*, 2000), ignoring the inter- and intra-individual factors that may affect transfer.

The PUF roller transferable residue sampler was developed to simulate the pressure applied to a surface by a crawling child weighing 9 kg (7,300 Pa) (Hsu *et al.,* 1990). The PUF roller consists of a weighted roller fitted with a thick, moistened polyurethane foam (PUF) cover. Modifications include using either a dry PUF roller cover or a thinner PUF skin. More details can be found in the literature (Hsu *et al.,* 1990; Lewis *et al.*, 1994; Stout and Mason, 2003).

Discussion of the CTEPP surface wipe samples is included here rather than in Section 4.3 because of the small volume (only 2 mL) of isopropyl alcohol used. Also, it should be restated that in CTEPP transferable residue samples were only collected in those homes and daycare centers that reported recent pesticide use.

Limits of detection for each method and chemical are given by study above in Table 4.2. Detection frequencies are given in Figure 4.11. The C18 Press and PUF roller results from Daycare are not included (or further discussed) due to extremely poor detection frequencies, with only one C18 and two PUF samples above the limit of detection.

Pesticide Presence in Transferable Residues

- Overall, the detection frequencies for transferable residues were substantially lower than those for total available residues.
- Chlorpyrifos was detected in greater than 75% of transferable residues in all of the studies except MNCPES.
- *Cis*- and *trans*-permethrin were detected in greater than 50% of the transferable residue samples collected in CTEPP. These measurements were made in a subset of homes with recent indoor applications of unidentified pesticides.
- Transferable residues were rarely detected in field studies by the modified C18 surface press sampler. In CHAMACOS, the detection frequency for chlorpyrifos was zero. In MNCPES, the detection frequencies on the floor and on other surfaces were 8 and 5 percent, respectively. The only exception was the DIYC study, where the post-application detection frequency for diazinon was greater than 50%.
- The modified C18 press sampler was more successfully used in the laboratory studies (Test House and Food Transfer studies) where residues were measured on all surface types sampled.
- CTEPP used IPA wipes with only 2 mL isopropanol instead of the 10 to 20 mL often applied for total available residue measurements. It is likely that the amount of pesticide residue recovered from the sampled surfaces is influenced by the amount of IPA applied to the wipe. Other variables that should be considered include location sampled within the room and last known pesticide application.

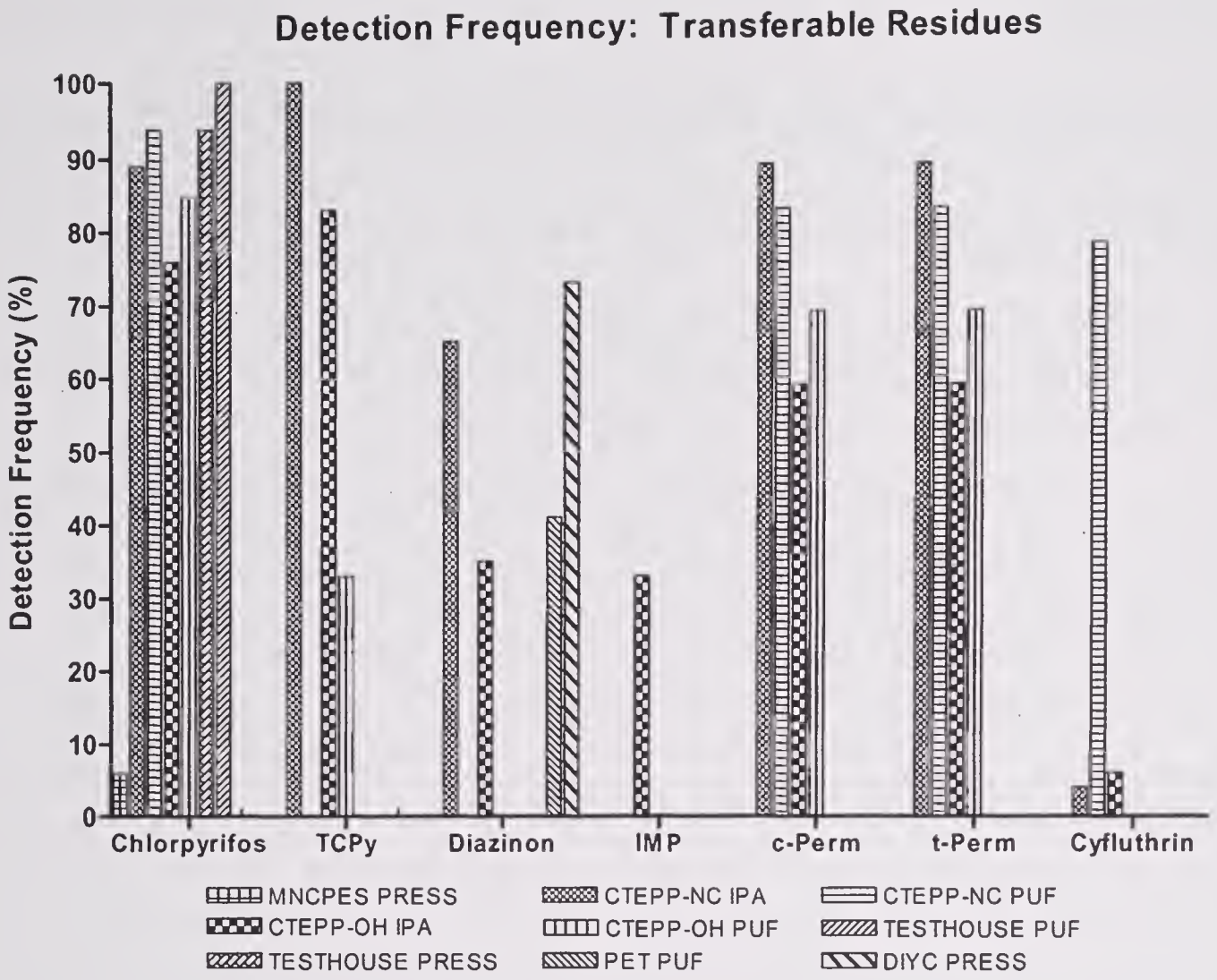


Figure 4.11 Detection frequencies for pesticides using transferable residue collection methods. All results from the C18 Press samplers used in CHAMACOS were below the limits of detection.

Transferable Residues: Summary Findings

Transferable residue loadings at the median and 95th percentile are given in Table 4.5 for all of the pesticides that were detected across studies (complete summary statistics are listed in Tables A.25 through A.29 in Appendix A). Transferable residue loadings of chlorpyrifos, diazinon, and permethrin are depicted in lognormal probability plots and box-and-whisker plots in Figures 4.12 and 4.13, respectively.

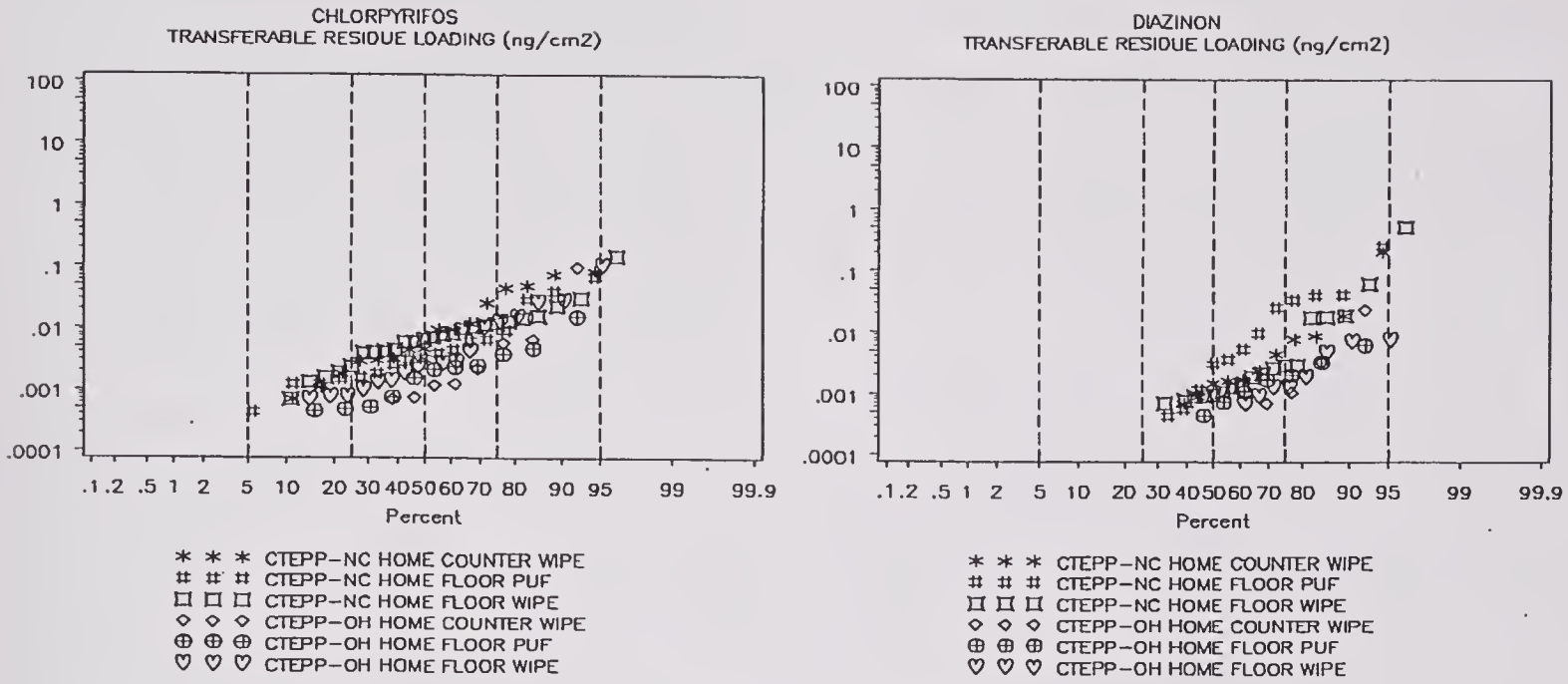
- The original C18 press sampler was designed to represent what adheres to the skin from a single hand press onto a carpeted surface. The uses for the modified C18 surface press sampler have expanded to include hard surfaces and longer contact times, contrary to its intended use. The data in Table 4.5 suggest that the sensitivity of the modified C18 surface press sampler is not adequate to measure typical residential pesticide residue levels due to its low collection efficiency (estimated as less than 1%).
- The mean transferable (2 mL IPA wipe) loadings were significantly different between CTEPP NC and OH for *cis*-permethrin ($p<0.01$), *trans*-permethrin ($p<0.05$), and diazinon ($p<0.01$). The mean loadings were not significantly different for either chlorpyrifos (ANOVA, $p=0.12$) or cyfluthrin (ANOVA, $p=0.17$).
- Wipe sampling methods varied in the volume of IPA used as a solvent (Table 4.1). The 2-mL IPA wipes used in CTEPP produced surface loading values that were very similar to those produced with the PUF roller (Figure 4.13). Since the PUF roller is a *transferable residue* method, it appears that the amount of IPA applied to the wipe determines the type of surface residue collected (*i.e.*, total or transferable residue). Interpretation of these results is complicated by other factors including recent application and sampling location with respect to application.

Table 4.5 Median and 95th percentile values for transferable residues (ng/cm²) by study.

Study	Method	Chlorpyrifos		Diazinon		*c*-Permethrin		*t*-Permethrin		Cyfluthrin		TCPy		IMP	
		P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
MNCPES	Press	<0.330	0.420	<0.140	1.13	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h [a]	IPA Wipe	0.007	0.140	0.001	0.51	0.050	1.500	0.034	1.600	<0.007	<0.007	0.005	0.024		
CTEPP-OH h [a]	IPA Wipe	0.002	0.760	<0.001	0.05	0.005	0.780	0.005	0.790	<0.007	0.041	0.001	0.033	<0.001	0.007
TESTHOUSE	PUF	0.005	0.15	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE	Press	0.230	6.90	--	--	--	--	--	--	--	--	--	--	--	--
PET	PUF	--	--	<0.005		--	--	--	--	--	--	--	--	--	--
DIYC	Press	--	--	3.80	24.0	--	--	--	--	--	--	--	--	--	--

--, pesticide not measured

[a] Homes only (daycares excluded)



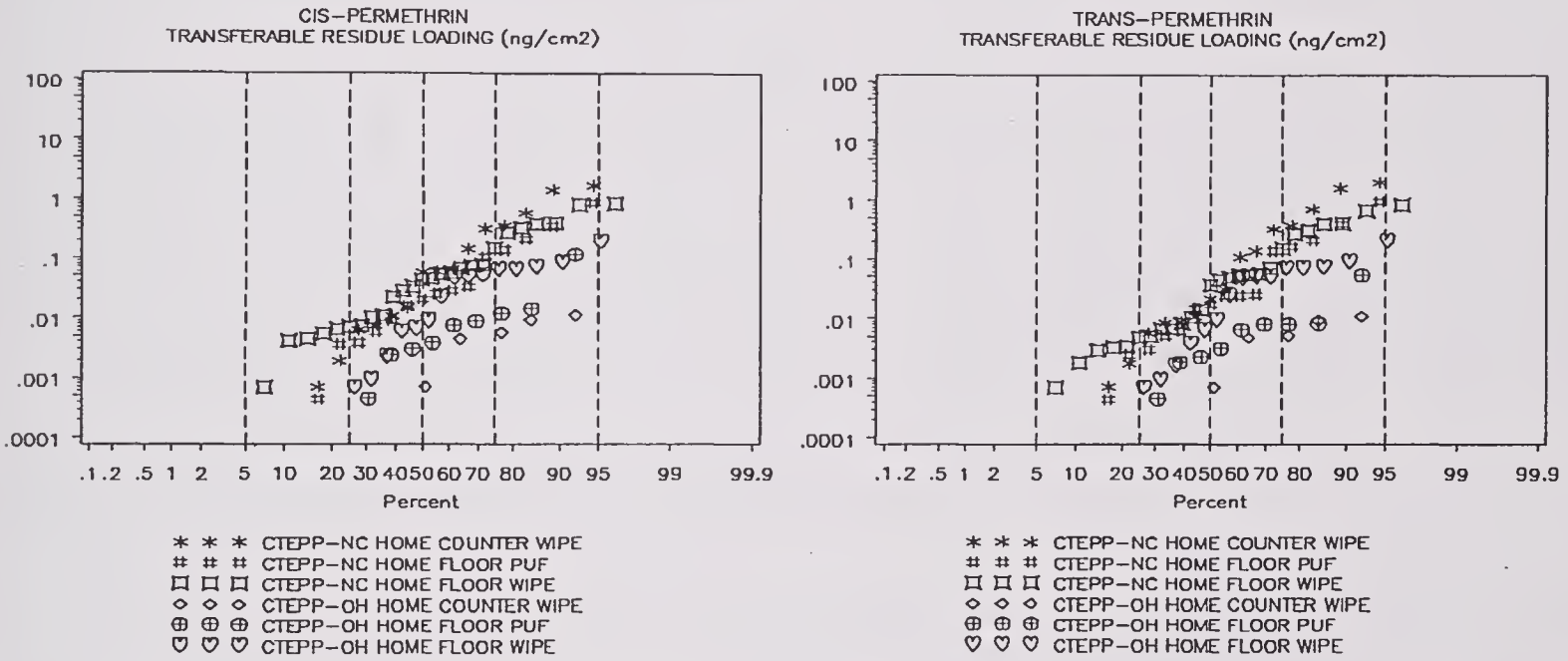


Figure 4.12 Lognormal probability plots for transferable residue loadings for the most frequently detected pesticides which include chlorpyrifos, diazinon, and *cis*- and *trans*-permethrin from CTEPP.

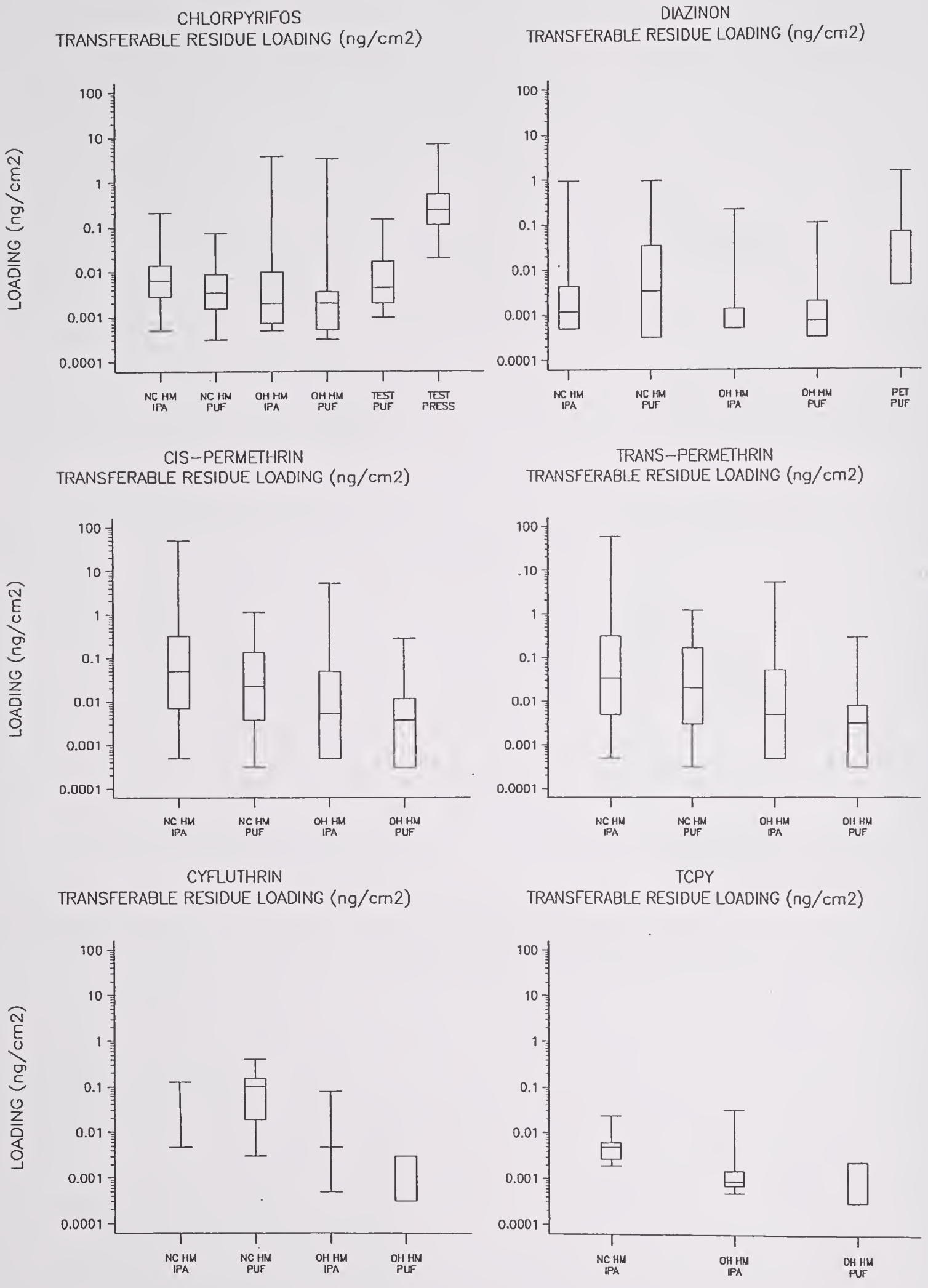


Figure 4.13 Box-and-whisker plots for transferable residue loadings for the most frequently detected pesticides which include chlorpyrifos, diazinon, *cis*- and *trans*-permethrin, cyfluthrin, and TCPy.

4.5 Spatial and Temporal Variability

Spatial and temporal variability were investigated in studies involving recent pesticide applications, including:

- Test House using IPA wipes, deposition coupons, C18 press sampler and PUF roller;
- CPPAES using IPA wipes, deposition coupons, and the LWW sampler;
- DIYC using IPA wipes and C18 press; and
- Daycare study using the IPA wipes.

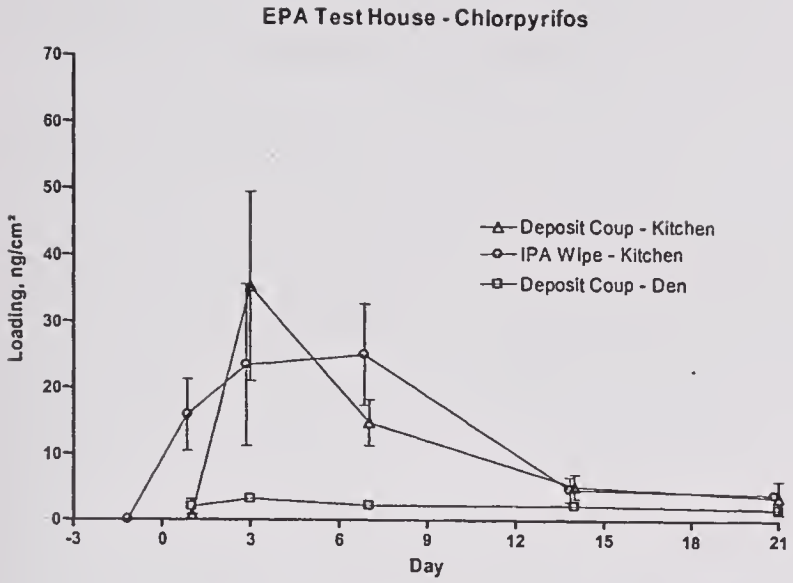
In studies with a series of measurements over time, the interval of time between measurements ranged from one to three days. In CPPAES, multiple rooms in ten homes were monitored for two weeks post application. In DIYC, multiple surfaces in three homes were monitored for one week. In the Test House, multiple surfaces in multiple rooms of a single house were monitored for 21 days. The Daycare study included multiple applications, each separated by one to three months, in a single daycare facility. In addition to sampling main activity areas, some studies also sampled less frequently contacted areas.

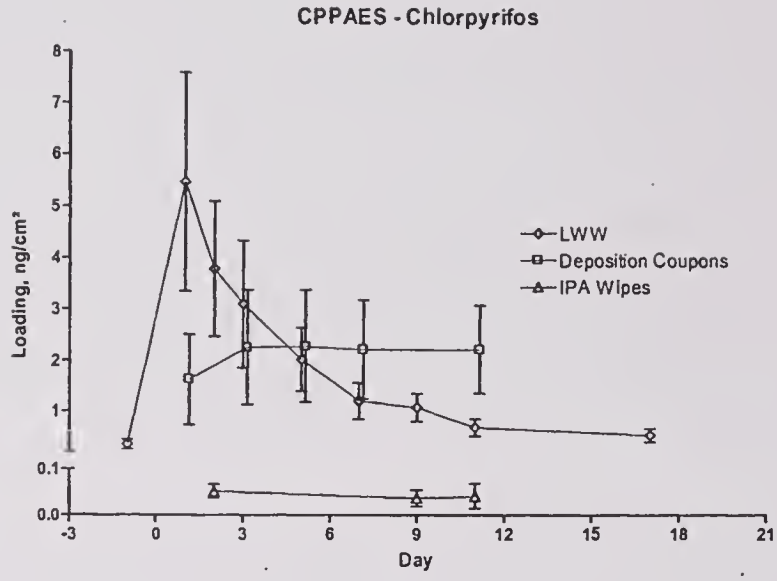
Figure 4.14 presents total available surface residue loadings measured in multiple locations in multiple rooms over time in the Test House, in multiple rooms in ten homes in CPPAES, and on multiple surfaces in three homes in DIYC. Figure 4.15 presents transferable residue measurements over time in multiple rooms of the Test House and on multiple surfaces in three homes in DIYC. Figure 4.16 presents total available residue measurements from the Daycare study, collected immediately following applications on multiple surfaces in two rooms. Figure 4.17 presents spatial variability in deposition coupon loadings in the kitchen (application site) and den (adjoining room) of the Test House following pesticide application.

Spatial and Temporal Variability: Summary Findings

- Preliminary examination indicates that total available residue loadings decay at a slower rate than airborne concentrations (See Figures 4.14 and 3.8).
- In the Test House experiment, the transferable residue loadings appeared to decrease at a faster rate than the total available residues (Figures 4.14 and 4.15). This may have occurred because the pesticide residue became less available for transfer (for example, due to an interaction with the surface or because the dried residue was less available for transfer).
- The transferable residues on the counters in DIYC (Figure 4.15) are nearly as high as those on the floors immediately after application, suggesting translocation of the pesticide from the site of application (assuming counters were not application surfaces).
- Substantial variability within rooms (at times a 100-fold difference in loadings) is evident in the Daycare data (Figure 4.16). Exposure estimates using measurements at a single location based on an assumption of homogenous surface loadings may result in exposure misclassification. The spatial variability points to the need for sampling of multiple locations and perhaps for better resolution in the activity data that is gathered.

- Data from the Test House (Figure 4.17) show that surface loadings cannot be assumed to be homogenous within a room.
- In the CCC study, loadings on floors were generally higher than loadings on table tops.
- In a published analysis of the MNCPES LWW wipe data, Lioy and colleagues (2000) reported substantial variability in surface chlorpyrifos levels among different rooms.





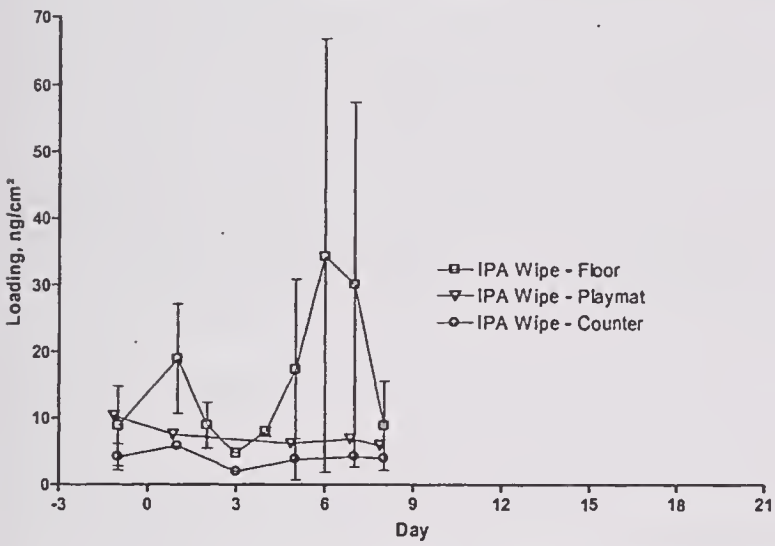
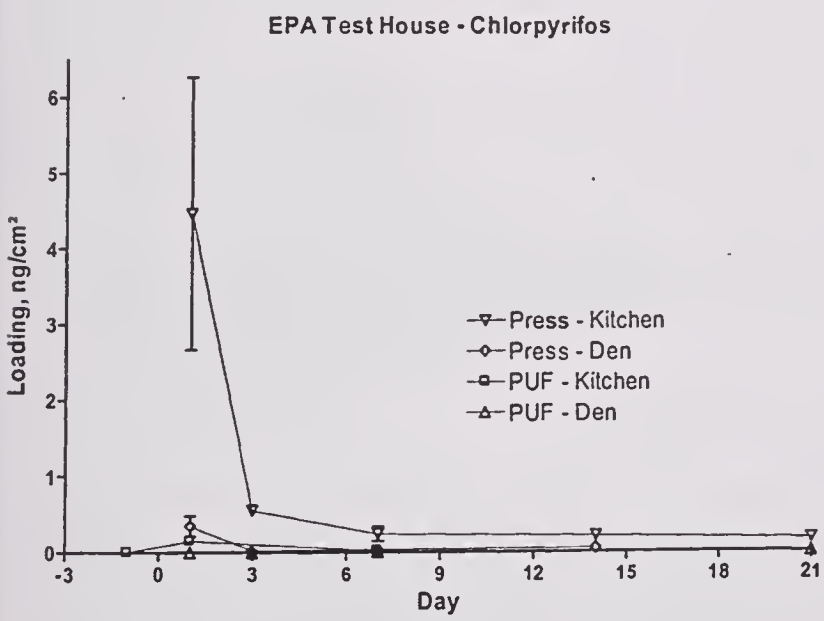


Figure 4.14 Total available surface residue loadings measured in multiple rooms over time in the Test House, in multiple rooms in ten homes in CPPAES, and on multiple surfaces in three homes in DIYC.



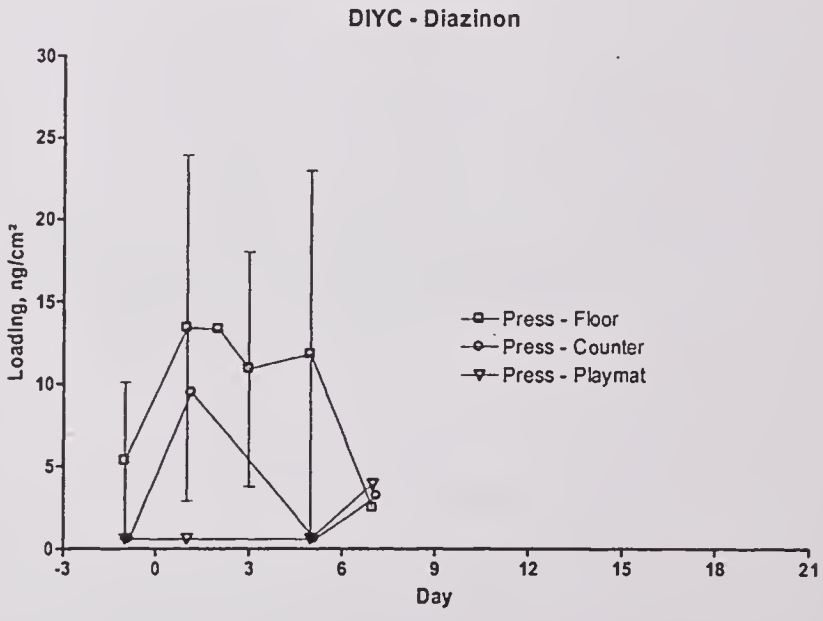


Figure 4.15 Transferable residue measurements over time following an application from multiple locations in multiple rooms of the Test House and multiple surfaces in three homes in DIYC.

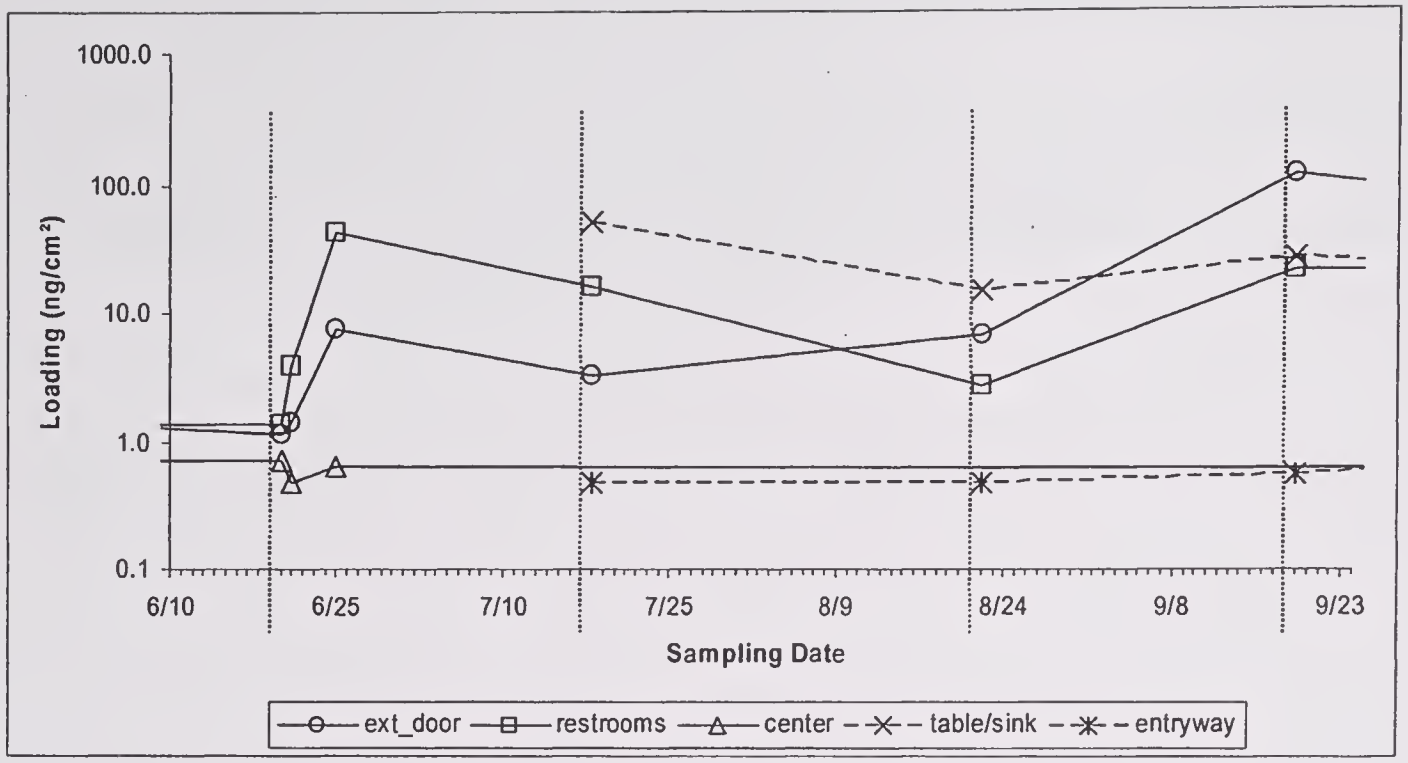


Figure 4.16 Total available residue measurements from the Daycare study, collected immediately following applications on multiple surfaces in two rooms in a single daycare facility. Solid Line represents the preschool room and dashed line represents infant room
Dotted vertical line represents application.

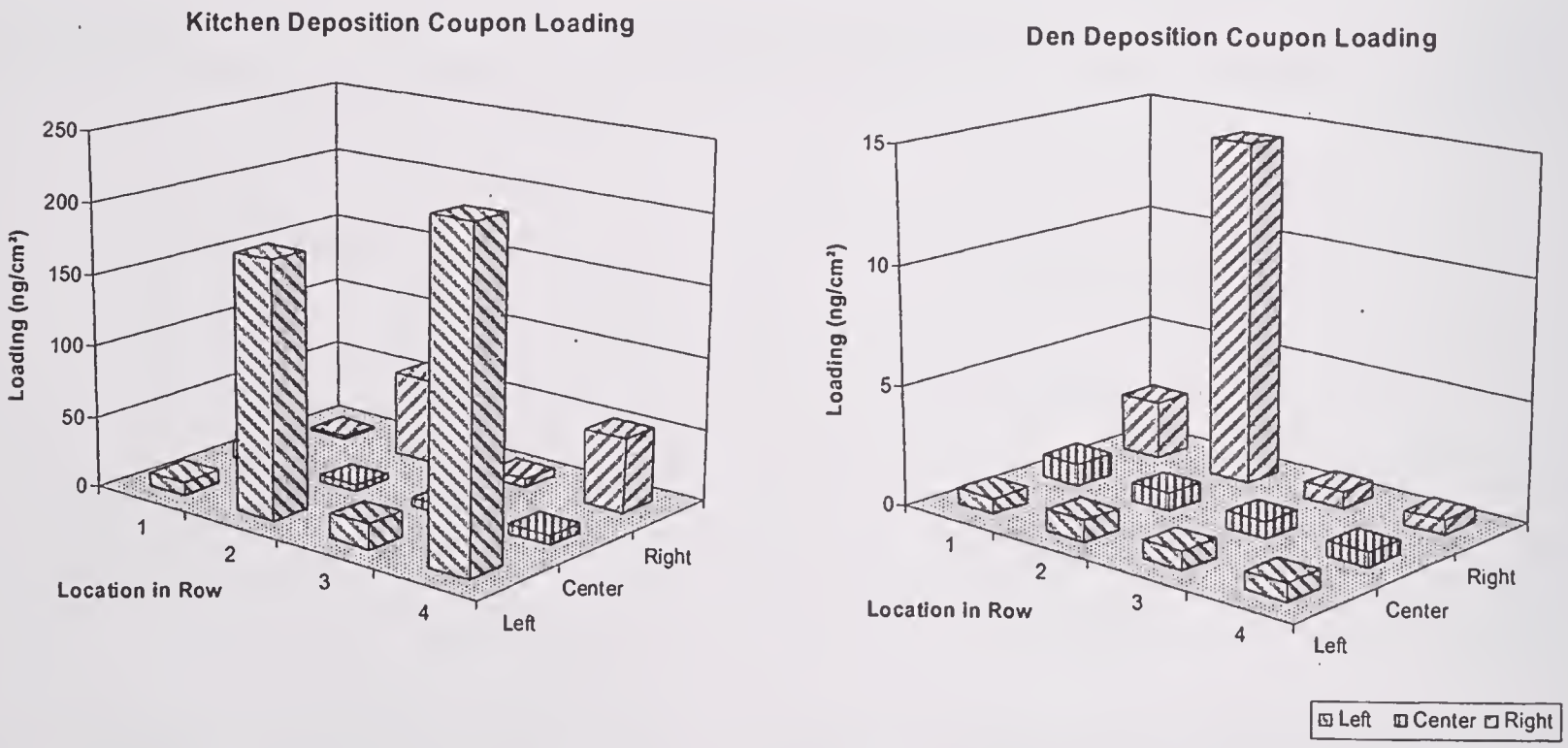


Figure 4.17 Spatial variability in deposition coupon loadings in the kitchen (application site) and den (adjoining room) of Test House following pesticide application.

4.6 Differences Related to Location

Regional Differences

Studies dating back to the Non-Occupational Pesticide Exposure Study (NOPES) from 1986 to 1988 (Whitmore *et al.,* 1994) have reported regional differences in environmental pesticide concentrations and loadings. Differences are thought to result from heavier use of insecticides in warm weather climates with higher year round insect control problems than in colder regions where hard winters help to curb insect populations.

- Median diazinon surface dust loadings (ng/cm^2) in home environments (daycares excluded) were very similar (about 0.002 ng/cm^2) across three states (NC, OH, and AZ, Table 4.3), and the 95th percentiles were also somewhat similar (0.12, 0.31, and 0.18, respectively). ANOVA analysis with Bonferroni adjustment for multiple comparisons found no significant differences among the three locations. These dust measurements do not provide evidence of the geographic variations consistent with geographic differences in pest treatment practices reported by Colt (1998).
- The overlapping distributions of pesticide concentrations in dust (ng/g) in the large observational field studies in Arizona, North Carolina, and Ohio (Figure 4.4) suggest that concentrations in dust may not be useful for determining region-specific pesticide use.
- For transferable residues obtained with 2-mL IPA surface wipes, the mean chlorpyrifos and cyfluthrin loadings were higher for CTEPP-NC compared to CTEPP-OH but not statistically different (Figures 4.12, 4.13). However, the mean loadings were significantly higher in NC for *cis*-permethrin (ANOVA; $p<0.01$) and *trans*-permethrin (ANOVA; $p<0.05$) and marginally significant for diazinon (ANOVA; $p<0.10$).
- Analysis of surface wipe samples from the national, probability-based Child Care Center study indicated no differences in the mean pesticide loadings among daycares in the four Census regions (data not shown, Tulve *et al.*, 2006).
- Differences in surface sampling methods, year of the study, and time of year when samples were collected make it difficult to examine any regional differences in surface pesticide loadings in homes. The transferable residue measurements suggest higher levels in NC than in OH, but no systematic differences are evident in dust concentrations or total surface residue loadings, although JAX had much higher surface loadings than any of the other studies without recent applications.

Urban vs. Rural

Lu and colleagues (2004) recently reported that at least one organophosphate pesticide was present in the house dust of 75% of agricultural area homes but only 7% of metropolitan area homes, suggesting different exposure pathways for children living in agricultural and nonagricultural regions. While concerns about pesticides may be more obvious in farming and other rural areas, widespread elevated pesticide residue levels have also been reported in highly urbanized minority communities of New York City (Whyatt *et al.,* 2002).

- Neither the median nor 95th percentile concentrations of chlorpyrifos measured in CHAMACOS dust was substantially higher than the median and 95th percentile in the other studies (Table 4.3). The assumption that children living in agricultural areas experience higher exposures than children in nonagricultural regions is not supported by these chlorpyrifos in dust measurements.
- Relatively high pre-application surface loadings in some of the CPPAES homes (data not presented) suggest possible contamination from pesticides applied in neighboring apartments in close proximity (Hore, 2003). Alternatively, the high loadings may suggest frequent treatments in those homes.

4.7 Influential Factors

As discussed above, the following factors appear to influence measured surface concentration or loading values:

Collection Methods

- The different types of collection methods are intended to have different collection efficiencies to serve different purposes. Efficiencies for various methods have been previously published.
- Total residue methods (which use both solvent and mechanical action to remove residues that may have penetrated into the surface) produce the highest values, followed by dust methods, and then by transferable residue methods.
- The low pesticide surface loadings obtained with 2 mL IPA wipes in both the NC and OH CTEPP studies (comparable to loadings obtained with the PUF roller) suggest that the amount of IPA applied to the wipe affects the amount of pesticide residue recovered.
- The C18 Press does not appear to be useful for determining typical surface pesticide residue loadings, for which it was never intended, because of its low collection efficiency and small size.

Surface Types

- Surface type has been shown to affect the collection efficiency of wipes. Recently published NERL data (Rohrer *et al.,* 2003) found that wiping from hard surfaces greatly exceeded carpet, and tile generally exceeded hardwood. As stated by Rohrer, "Highest pesticide recoveries were from tile with diazinon (59%), chlorpyrifos (80%), and permethrins (52% *cis*; 53% *trans*) being the only pesticides recovered by wiping at greater than 50% of the applied concentrations."

Sampling Locations

- Despite evidence of translocation from direct application areas, the application area surface residue loadings were generally higher than the play area surface residue loadings in JAX.

- In the CCC study, floor residue loadings were typically higher than table top or desk top loadings.
- Experiments in the Test House showed high spatial variability in loadings in the room of application (kitchen) and transport of pesticide residues to the adjoining room.
- Results from the Daycare study showed substantial differences in surface loadings (up to two orders of magnitude) at different locations in a daycare center.

Occupant Activities

- Surface chlorpyrifos loadings were reportedly lower in the CPPAES homes in which the occupants performed cleaning activities and/or the homes that had high ventilation rates (Hore, 2003).
- Crack and crevice applications in the unoccupied Test House produced higher surface loadings and longer decay times than the same type of application (albeit with less active ingredient released) in the occupied CPPAES homes.

Pesticide Use Patterns

- On a regional level, surface loadings in Jacksonville, Florida, an area likely to have year-round pest control issues and high pesticide usage, were much higher than in any of the other observational studies.
- Within a given region, however, pesticide use information collected with questionnaires or inventories may not correlate with measured surface values. Published results from the MNCPES indicate that the residential pesticide use questions and overall screening approach used in the MNCPES were ineffective for identifying households with higher levels of individual target pesticides (Sexton *et al.*, 2003).

4.8 Correlations among Soil, Wipes, and Dust

- Analysis of CCC data (Tulve *et al.*, 2006) found little correlation between surface wipe loadings and soil concentrations for 16 common organophosphate and pyrethroid pesticides.
- In the CTEPP study, significant Spearman correlations between dust and soil concentrations were observed with diazinon ($r=0.26$, $p<0.01$) and TCPy ($r=0.21$, $p<0.05$) in NC homes and chlorpyrifos ($r=0.28$, $p<0.01$) and TCPy ($r=0.20$, $p<0.05$) in OH homes (data not presented).
- Identification of correlations is hindered by the low detection frequencies for many pesticides in soil.

4.9 Particle-Bound Pyrethroid Residues: Implications toward Exposure

The recent shift in commonly applied residential pesticides from organophosphate to pyrethroid compounds carries with it important implications for human exposure. The chemical and physical properties of a pesticide govern its behavior with respect to movement and fate. In general, pyrethroids have properties that favor the particulate phase, resulting in transport mechanisms preferentially involving dust rather than vapor. A tendency towards the particulate phase also suggests a decreased relative importance of the inhalation route and an increased relative importance of the dermal and indirect ingestion routes.

Pesticides applied in homes translocate from the point of application and deposit onto non-target surfaces. Because human contact with target surfaces (*e.g.*, cracks and crevices) is typically obstructed or otherwise hindered, it is largely the movement of residues from the point of application into the air and onto non-target surfaces that results in exposure. The movement of residentially applied insecticides follows a complex and poorly understood process of transformation and phase distribution and is influenced by several factors, namely: delivery system, application surface type, solvent, formulation, physicochemical properties of the active insecticide, and human and companion animal activity.

Overall, pyrethroids have similar physicochemical properties, and as a result, they display similar behavior in the residential environment (Laskowski, 2002; Oros and Werner, 2005). Pyrethroids generally have low vapor pressures and Henry's Law constants, thus they resist volatilization and exist almost entirely in the particulate phase at room temperature. They have high octanol/water partition coefficients (K_{ow}), which suggests they tend to partition into lipids, and very high water/organic carbon partition coefficients (K_{oc}), which suggests that they also tend to partition into organic matter. With these characteristics, pyrethroids can be expected to bind readily to the particulate matter that comprises house dust. Particles resuspended by human activity then act as the primary vector for pyrethroid transport and for human exposure.

Particle-phase contaminant transfer is strongly particle size dependent (Rodes *et al.*, 2001). Kissel *et al.* (1996) reported that dermal adherence of dry soil primarily involves particles in the <150 μm size fraction. Assuming that house dust behaves similarly with respect to transfer, the size fraction that preferentially adheres to skin not only comprises the bulk of house dust, but also contains the highest pesticide concentrations. Rodes *et al.* (2001) reported that the <150 μm size fraction comprises about 60% of house dust. Pesticide concentrations in house dust increase with decreasing particle size, and are highest in the <25 μm size fraction (Lewis *et al.*, 1999). Because the surface-to-volume ratio similarly increases with decreasing particle size, pesticides appear to be primarily attached to the surfaces of the particles (rather than trapped within).

Particle-bound movement and transfer of pyrethroids imply a decreased importance of the inhalation route and an increased importance of the indirect ingestion route. Exposure of young children, for whom indirect ingestion of residues from object- and hand-to-mouth activities is particularly important, may be most strongly affected. Particle-bound residues may also have a reduced potential for dermal absorption, as a consequence of being bound to the particle.

5.0 DIETARY EXPOSURE MEASUREMENTS

5.1 Introduction and Data Availability

Diet can be a significant pathway of exposure to humans. Infants and young children may be particularly vulnerable to exposure by dietary ingestion because they eat more than adults do relative to their body weights. Foods may contain residues of pesticides because of intentional agricultural applications or they may become contaminated during processing, distribution, storage, preparation, and even consumption. The ingestion of residues on foods resulting from contact with hands and surfaces during consumption as well as the ingestion of pesticide residues while mouthing contaminated hands and objects are considered "indirect ingestion" pathways and are the subject of the next chapter (Chapter 6.0). This chapter provides a comparative summary of measurements of pesticides in duplicate diet samples and of estimated dietary intakes. The sample collection methods for the studies that included duplicate diet measurements are summarized in Table 5.1.

Among the large observational studies, duplicate diet samples were collected in NHEXAS-AZ, MNCPES, and CTEPP. In CTEPP, food and beverage samples were collected at both homes and daycares. Duplicate diet samples were also collected in three pilot-scale studies, CHAMACOS (20 participants), DIYC (three participants), and JAX (nine participants).

- The most common measure of dietary exposure was by composited duplicate diet analyses (Table 5.1). This approach reduces study costs compared to analyzing individual foods, but it increases the complexity of the sample analysis and produces higher method detection limits.
- Duplicate diet samples measure the pesticide residues in the children's foods after processing and preparation by the caregiver. The samples, therefore, may include residues from contaminated food handling surfaces in addition to the residues contained in the food products. However, duplicate diets fail to capture the additional intake of pesticides resulting from the child's activities before and during consumption, as discussed in Chapter 6.
- Duplicate plate samples were used for dietary measurements at the daycares in CTEPP. The distinction between a duplicate plate and a duplicate diet (with the latter accounting for uneaten foods) is typically more important for children than adults because significant quantities of food may be left uneaten.

Table 5.1 Dietary exposure sample collection methods for pesticides.

Study	Children Ages (years)	Sample Type	Collection after Indoor Pesticide Use	Mass Recorded	Collection Period	Sample Handling	Composite	Relevant Analytes
NHEXAS-AZ	6 - 12	Duplicate diet	No	No	24 hr	Liquid and solid food collected separately in polyethylene containers	Yes	Chlorpyrifos, diazinon
MNCPES	3 - 12	Duplicate diet	No	Yes	4 d	Liquid and solid food collected separately; solid food split into potentially "high pesticide" foods and "remaining" foods	Yes	Chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin
CTEPP	2 - 5	Duplicate diet (homes), and duplicate servings (at daycare centers)	No	Home samples only	48 hr	Liquid and solid food collected separately in glass jars	Yes	Chlorpyrifos, TCPy, diazinon, IMP (Ohio only)
JAX	4 - 6	Duplicate diet	Yes	Yes	24 hr	Solid and liquid food stored in polyethylene containers	Yes	Chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cyfluthrin
CHAMACOS	0.5 - 2	Duplicate Diet	No	Yes	24 hr	Liquid collected in polycarbonate bottles and solid food in polyethylene zip closure bags	Yes	Chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cyfluthrin
DIYC	1 - 3	Duplicate diet, each food collected individually	Yes	Yes	24 hr	Each food stored in individual zip-loc bags	No	Diazinon

5.2 Pesticide Presence

Table 5.2 presents the detection limits for the studies. The frequency of detection for the selected pesticides is presented in Figure 5.1. The median and 95th percentile concentrations are presented in Table 5.3. Data are presented in lognormal probability plots (Figures 5.2 and 5.3) for the large observational field studies and box-and-whisker plots (Figures 5.4 and 5.5) for all of the studies. Where food mass measurements are available (Table 5.1), both concentration and intake (mass of compound ingested) are presented. Intake is defined as µg/day in keeping with the dietary exposure algorithm of the *Draft Protocol* (Berry *et al.*, 2001) rather than as µg/kg-bw/day which would be more consistent with the reference dose (RfD) paradigm.

- Reported method detection limits for chlorpyrifos ranged from 0.04 µg/kg in JAX up to 1.7 µg/kg in CHAMACOS (Table 5.2).
- Chlorpyrifos was detected in over 50% of the duplicate diet samples in MNCPES, CTEPP, and JAX (Figure 5.1). The median chlorpyrifos concentrations in the MNCPES and JAX diet samples were at least twice as high as in the CTEPP samples (Table 5.3).
- Diazinon was not frequently detected in any of the studies except DIYC, a study in which there had been prior indoor applications. The data from DIYC suggest that contamination of food due to handling and surface contact is important in homes with recent applications (see Section 6).
- While detection of diazinon in food samples was typically below 30% (Figure 5.1), detection immediately following crack and crevice application in DIYC was 100%.
- The logplots (Figures 5.2 and 5.3) show that in the upper half of the distribution (between the 50th and the 95th percentiles), higher concentrations of *cis*- and *trans*-permethrin were measured in solid food in North Carolina homes than in North Carolina daycares or Ohio homes or daycares.
- Model simulations using DIYC data (results not presented) revealed that pesticides transferred to food during contact with surfaces and handling by a child may increase dietary intake significantly (over 60% under the modeled scenario).
- Published results from the MNCPES (Clayton *et al.*, 2003) showed that extant residue databases can successfully be used to select samples for analysis, potentially reducing costs by avoiding analyses of foods not likely to contain measurable levels. Care must be taken, however, to avoid neglecting those residues that are transferred during handling.
- Measurable levels of these particular pesticides were rarely detected in beverages in any of these studies. Future studies with other such pesticides that are not expected to be found in drinking water may consider eliminating this costly measurement.
- Infants and children consume far fewer types of foods than do adults (while consuming much more of certain foods) (NRC, 1993). Thus, the number of days of collection may be less important for children than for adults.
- The large potential for enzymatic degradation of pesticides (especially chlorpyrifos) during food sample storage and during homogenation prior to analysis has not been directly addressed by any studies under this program.

Table 5.2 Limits of detection (µg/kg) for pesticides measured in duplicate diets.

Study	Compounds				
	Chlorpyrifos	Diazinon	*cis*-Permethrin	*trans*-Permethrin	Cyfluthrin
NHEXAS-AZ	1.0	0.7	--[a]	--	--
MNCPES	0.26	0.3	0.2	0.2	--
CTEPP	0.08	0.08	0.08	0.08	0.83
JAX	0.04	0.04	0.02	0.02	0.4
CHAMACOS	1.4	1.2	4.5	2.9	--
DIYC	--	0.36 – 1.25	--	--	--

[a] Blank cells (--) indicate that the pesticide was not measured in the study.

Table 5.3 Median and 95th percentile pesticide concentrations (µg/kg) measured in duplicate diet food samples.

Study	Chlorpyrifos		Diazinon		*cis*-Permethrin		*trans*-Permethrin		Cyfluthrin	
	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
NHEXAS-AZ	BDL[a]	5.7	1.8	1.9	--[b]	--	--	--	--	--
MNCPES	0.53	2.4	BDL	0.38	--	--	--	--	--	--
CTEPP-NC Home	0.2	2.1	BDL	0.4	BDL	15.6	BDL	8.7	BDL	0.9
CTEPP-NC Daycare	0.1	0.9	BDL	0.2	BDL	5.2	BDL	3.0	BDL	BDL
CTEPP-OH Home	0.2	1.6	BDL	0.2	BDL	8.8	BDL	8.0	BDL	BDL
CTEPP-OH Daycare	0.1	0.6	BDL	0.2	BDL	2.2	BDL	1.4	BDL	BDL
JAX	0.38	7.4	BDL	1.0	0.29	13	0.22	22	BDL	3.6
CHAMACOS	BDL	1.4	BDL	BDL	BDL	BDL	BDL	BDL	--	--
DIYC	--	--	0.17	0.78	--	--	--	--	--	--

[a] BDL, Below minimum detection limit

[b] Blank cells (--) indicate the pesticide was not measured in the study

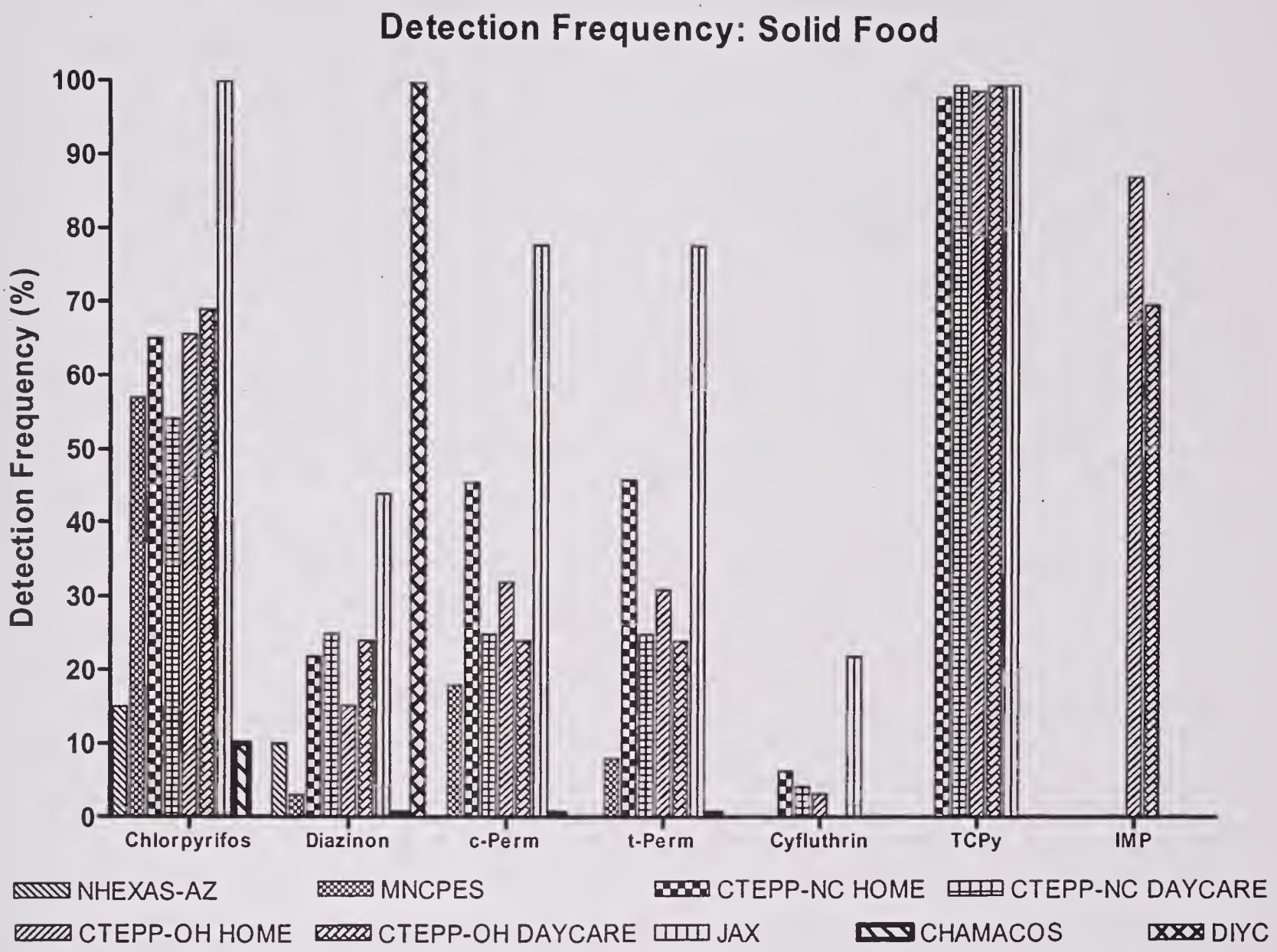


Figure 5.1 The detection frequency of pesticides measured in duplicate diet food samples.

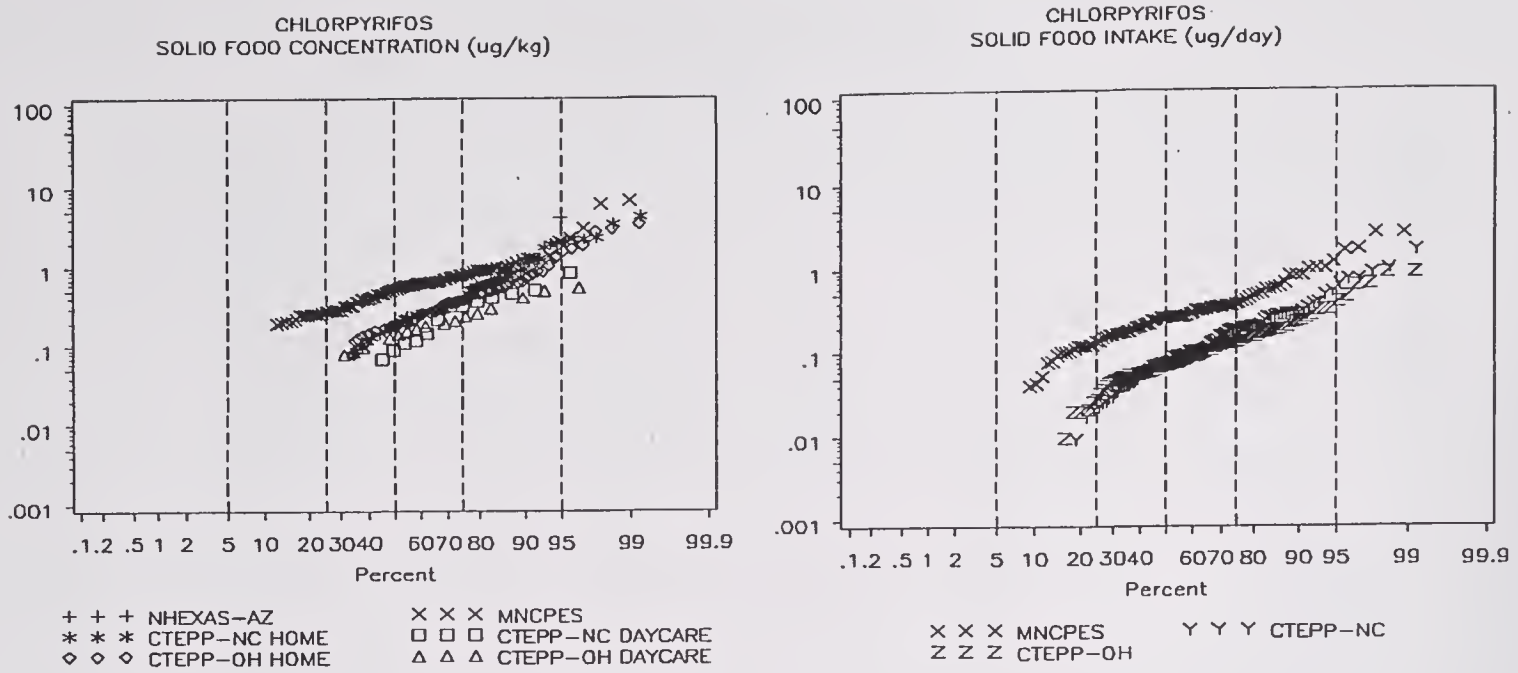
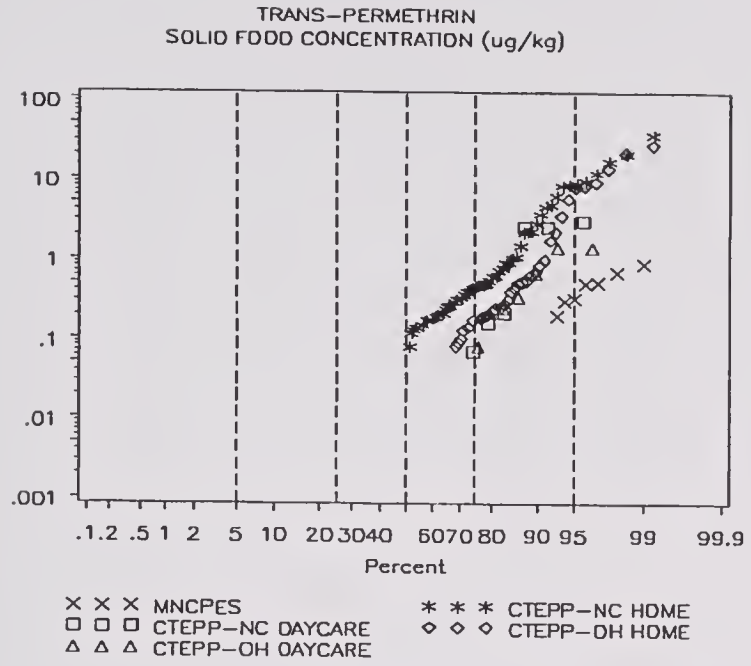
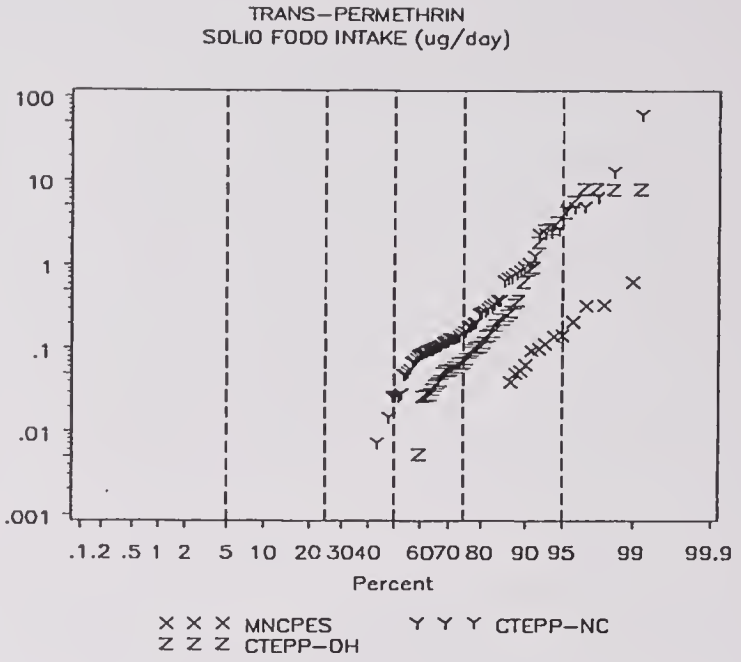
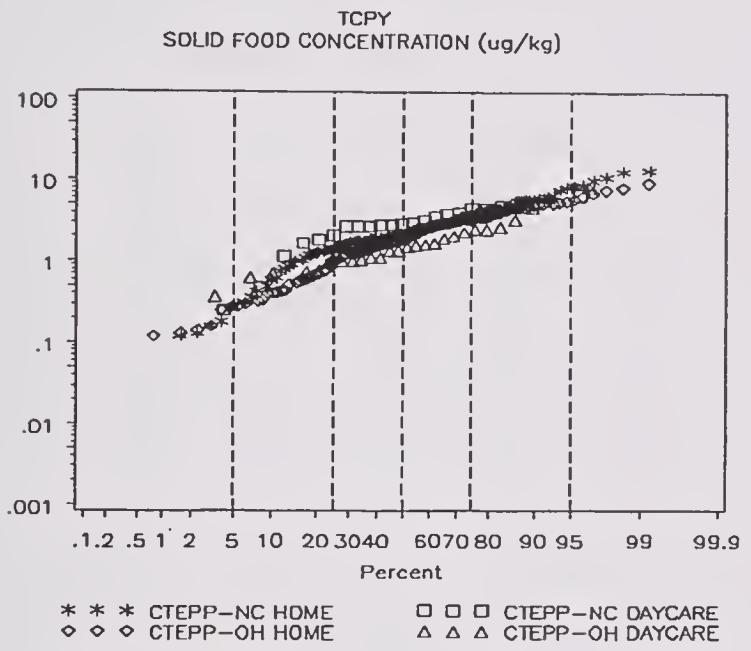
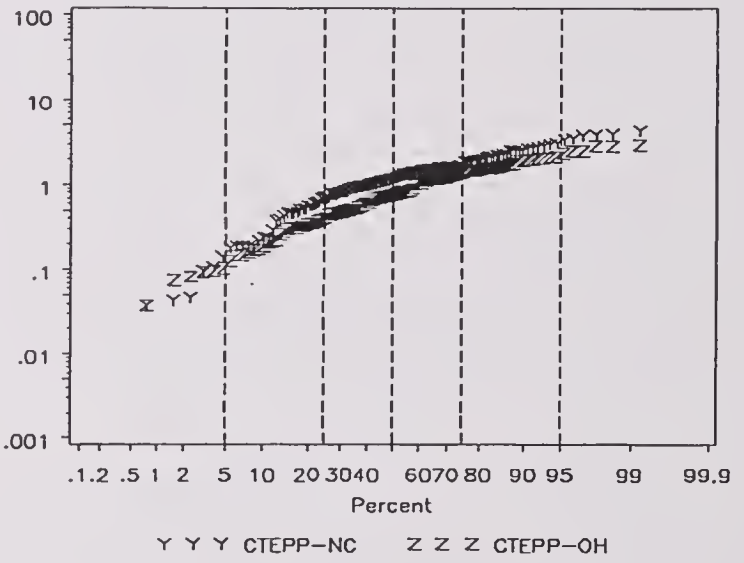


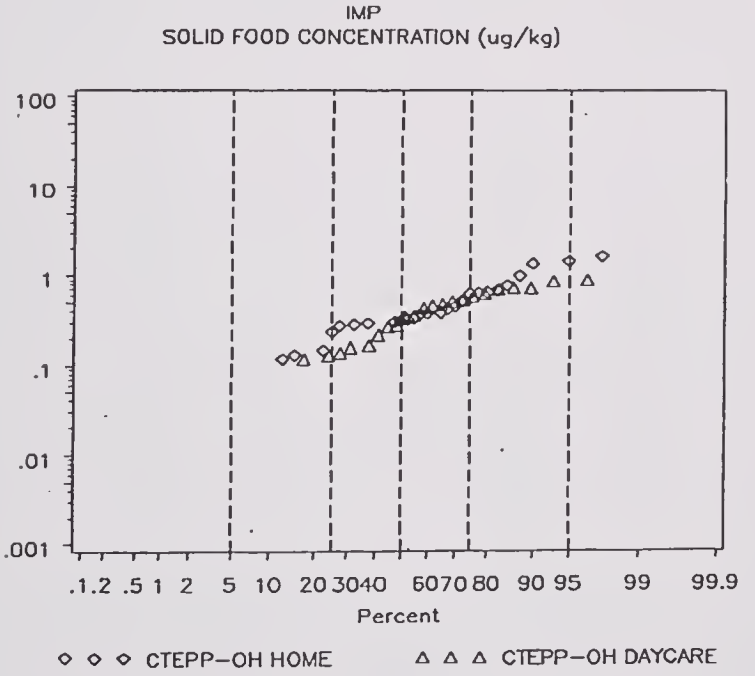
Figure 5.2 Lognormal probability plots of solid food concentrations (µg/kg) and intakes (µg/day) for chlorpyrifos, diazinon, and *cis*-permethrin from large observational field studies.











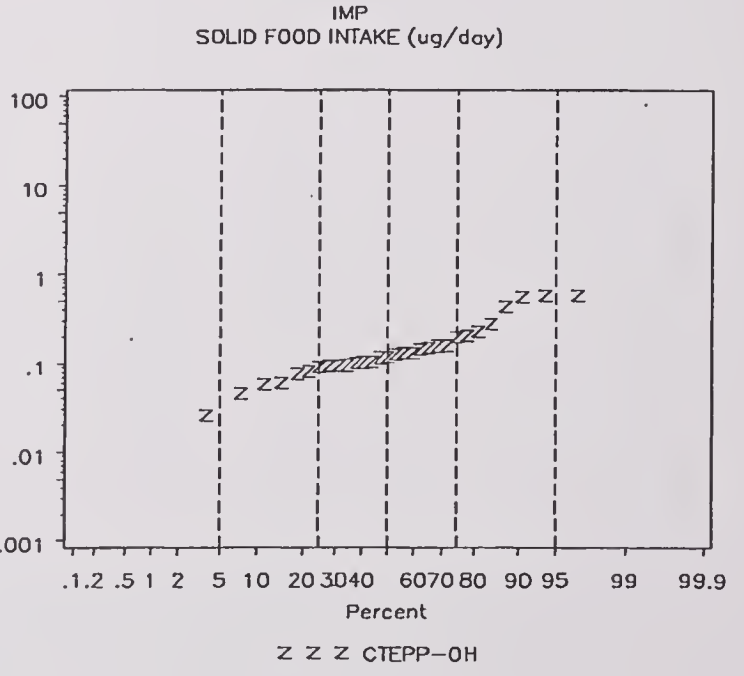


Figure 5.3 Lognormal probability plots of solid food concentrations (µg/kg) and intakes (µg/day) for *trans*-permethrin, TCPy, and IMP from large observational field studies.

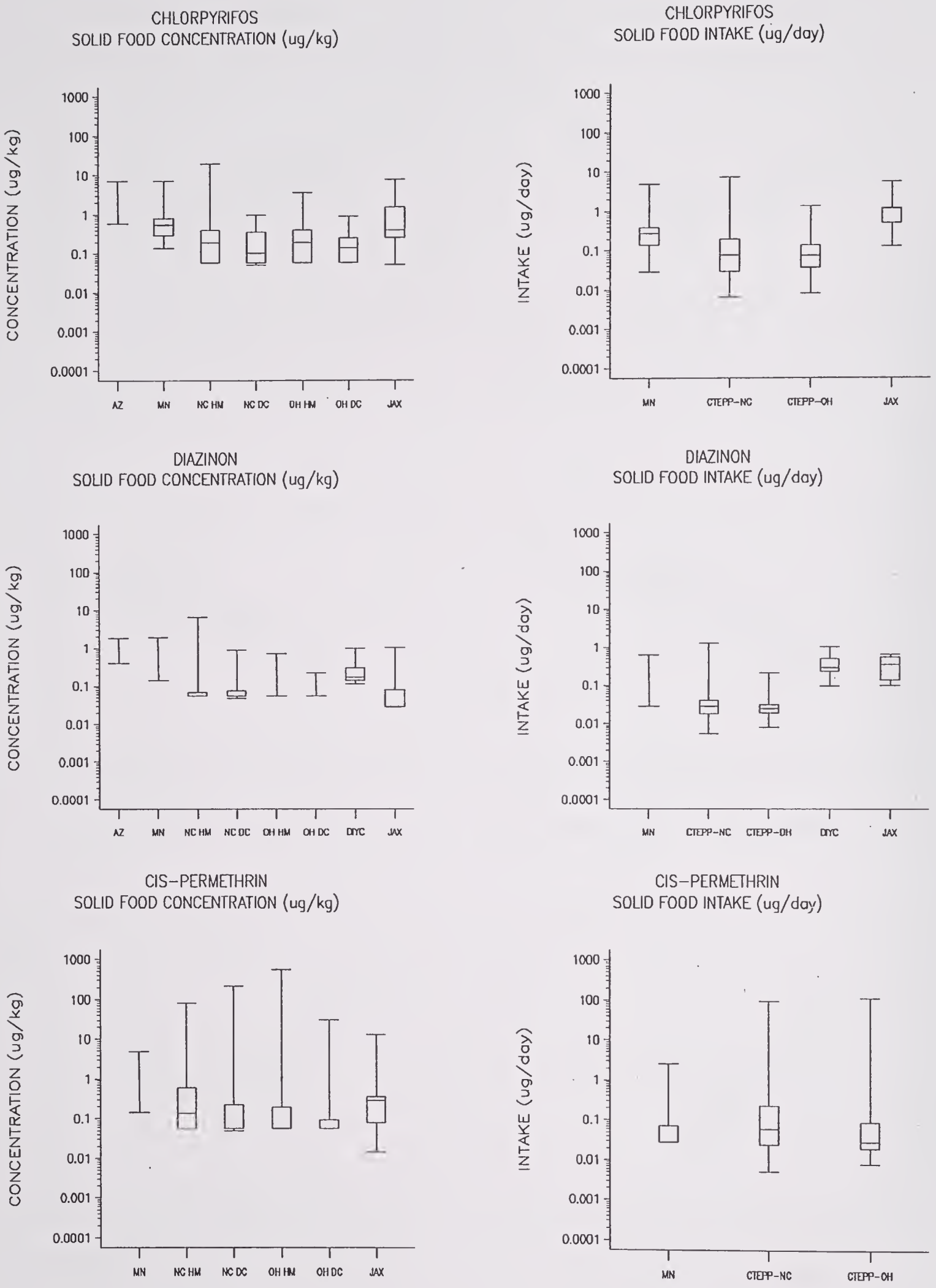


Figure 5.4 Box-and-whisker plots of solid food concentrations (µg/kg) and intakes (µg/day) for chlorpyrifos, diazinon, and *cis*-permethrin across all studies.

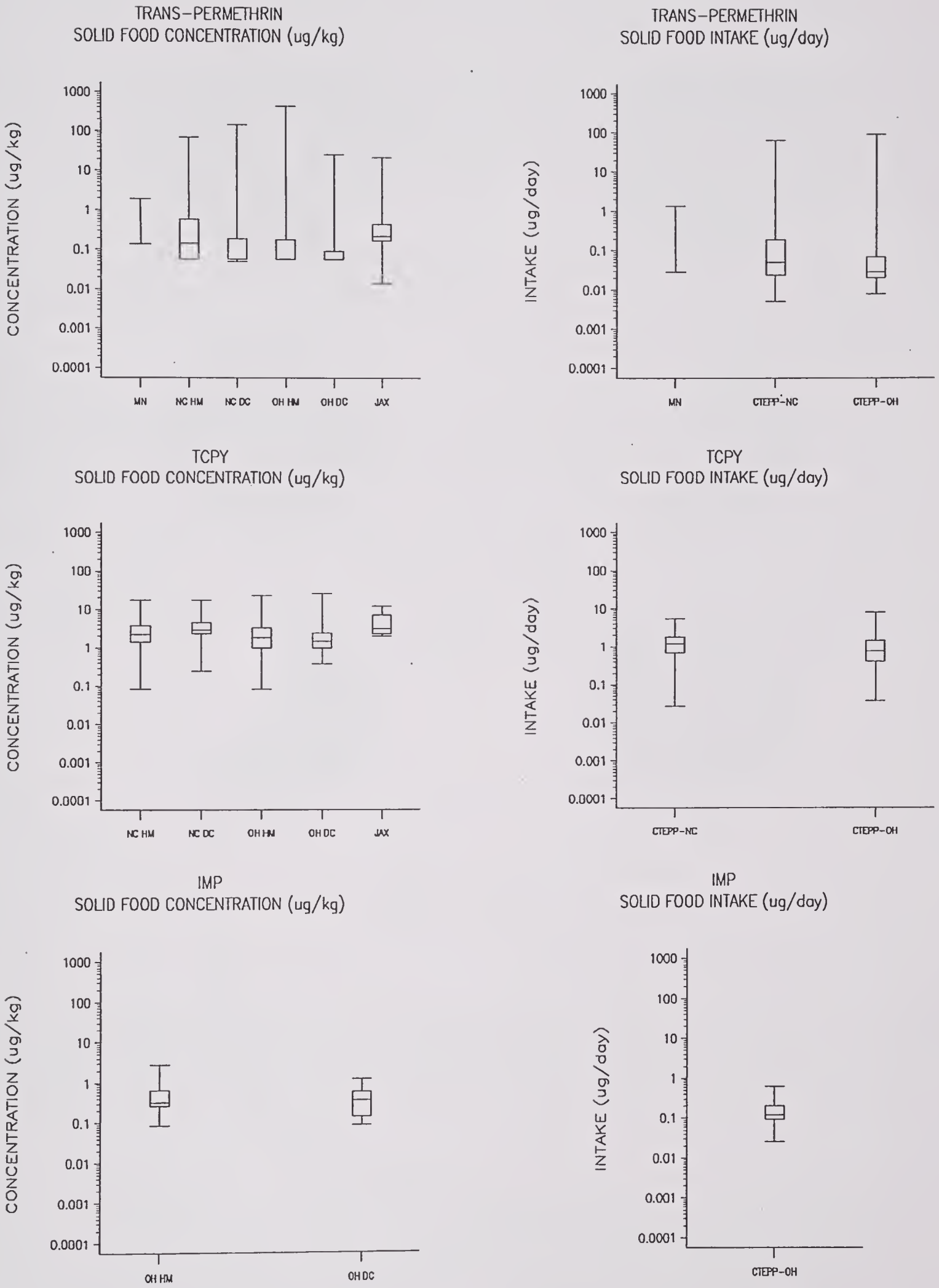


Figure 5.5 Box-and-whisker plots of solid food concentrations (µg/kg) and intakes (µg/day) for *trans*-permethrin, TCPy, and IMP across all studies.

5.3 Relative Importance of the Ingestion Route

The Stochastic Human Exposure and Dose Simulation (SHEDS) model (Zartarian *et al.*, 2000) prediction for dietary intake of *cis*-permethrin is compared to CTEPP measurements in Figure 5.6. The estimated proportion of aggregate exposure represented by dietary intake for CTEPP-NC and CTEPP-OH children is from the CTEPP Report (Morgan *et al.*, 2004) and is presented in Figures 5.6 and 5.7, respectively.

- An example of use of the SHEDS model to predict dietary intake of *cis*-permethrin in a study population is shown in Figure 5.6. The dietary intake estimates may then be compared to SHEDS model estimates of intake by other relevant routes to determine the relative importance of the ingestion route.
- Based on route-specific estimates (Figures 5.7 and 5.8), dietary ingestion represents the dominant route of exposure for chlorpyrifos, diazinon, and permethrin in the CTEPP study. Indirect ingestion, estimated based on dust and soil measurements, is a far greater concern for the permethrin than for chlorpyrifos and diazinon in the CTEPP study.
- The route that represents the dominant route of exposure (dietary ingestion) is also the route with the lowest detection frequencies (approximately 2/3 of the values for permethrin in CTEPP are nondetects), which increases the uncertainty in the estimates. Substituting a fraction of the detection limit for values below the limit of detection may have a disproportionate impact on the outcome.

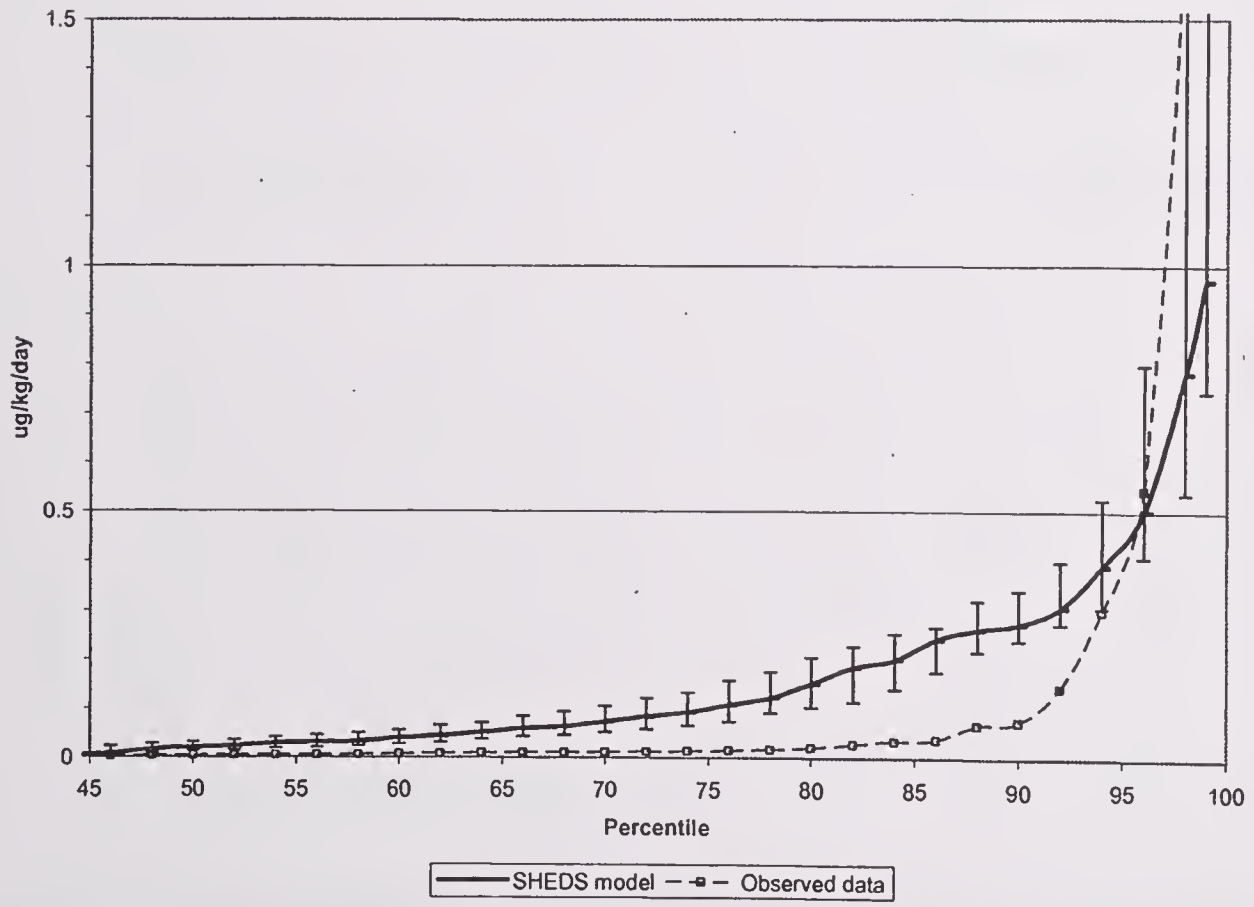


Figure 5.6 Comparison of SHEDS model prediction for dietary intake of *cis*-permethrin (µg/kg/day) and CTEPP measurement data.

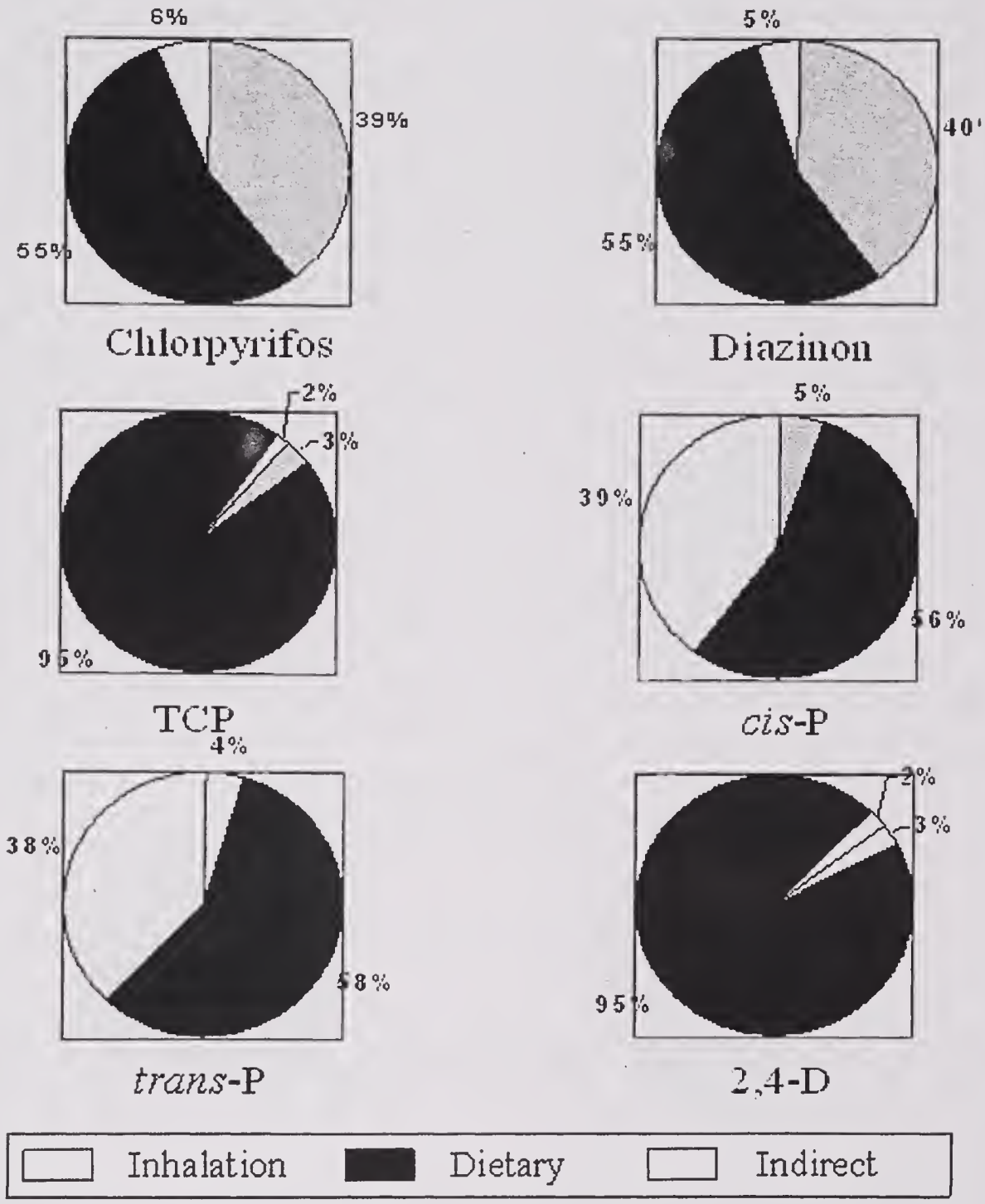


Figure 5.7 Estimated mean proportion of aggregate potential exposure for CTEPP-NC children by exposure route. (TCP = 3,5,6-Trichloro-2-pyridinol; *cis*-P and *trans*-P = *cis*- and *trans*-Permethrin; 2,4-D = 2,4-Dichlorophenoxyacetic acid.) From Morgan *et al.*, 2004.

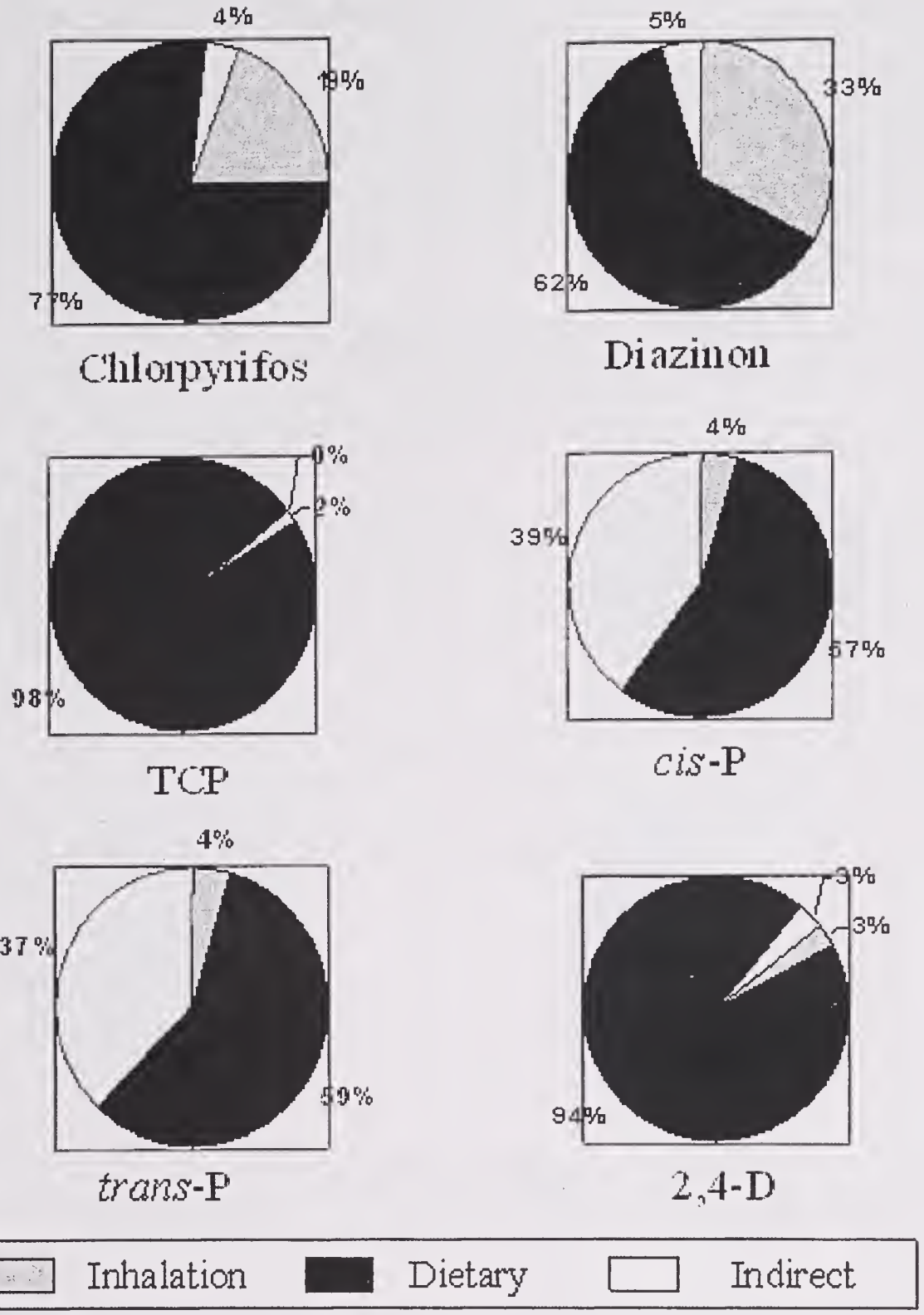


Figure 5.8 Estimated mean proportion of aggregated potential exposure for CTEPP-OH children by exposure route. (TCP = 3,5,6-Trichloro-2-pyridinol; *cis*-P and *trans*-P = *cis*- and *trans*-Permethrin; 2,4-D = 2,4-Dichlorophenoxyacetic acid.) From Morgan *et al.*, 2004.

6.0 INDIRECT INGESTION MEASUREMENTS

Children's ingestion of pesticide residues is not limited to residues in food and beverages acquired during cultivation, food production, and in-home preparation. Indirect ingestion refers to the ingestion of residues from hands or objects that enter the mouth, as well as to the ingestion of residues transferred to food items by contact with the floor or other contaminated surfaces during consumption. Indirect ingestion is believed to be an important route of exposure for children because of their frequent mouthing activities and their unique handling of foods while eating. Indirect ingestion may be the result of hand-to-mouth, object-to-mouth, or hand-to-object-to-mouth activity. Indirect ingestion may be estimated using an approach that lumps some of the exposure factors and activity patterns associated with indirect ingestion. This simplified approach allows for assessment of indirect ingestion exposure based on measurement data collected in the field and on factors that characterize the activities that lead to indirect ingestion. In this approach, objects (including food) that are commonly handled, mouthed, and/or ingested are identified in the field. The residue loadings on these objects are measured directly or estimated from surface loading measurements combined with transfer efficiencies measured in the laboratory. General information relating to the frequency and nature of these mouthing and ingestion activities is also collected. Data on the fraction of residues that may be removed from an object during mouthing that has been collected in the laboratory is then required to complete the assessment. In addition, the items identified as most often mouthed and/or eaten are assumed to represent the most significant sources of indirect ingestion exposure. This section presents summary data for studies addressing the indirect ingestion route of exposure (Table 6.1). Highlights of the data are presented below.

6.1 Characterizing Hand- and Object-to-Mouth Activities

Exposure models are based on two factors: how much pesticide residue is available for human uptake and what human activities occur that would result in contact with and uptake of residues. Hand-to-mouth and object-to-mouth activities are believed to directly impact ingestion of pesticides among children through the indirect ingestion exposure route, but the relative importance of these activities has not been established. In fact, the lack of empirical data showing that either hand- or object-to-mouth activities appreciably affect exposure makes it a hypothesis that has not yet been adequately addressed. The frequency of hand-to-mouth, object-to-mouth, and/or combo-to-mouth contacts were quantified for children in the MNCPES and CPPAES studies using a computer software system (Table 6.2). These studies used Virtual Timing Device (VTD) software (Zartarian *et al.*. 1997) to quantify the children's normal daily activities captured on videotape. The following are highlights of the data from these studies.

- Assigning contact as either a hand-to-mouth or an object-to-mouth contact can cause the hand-to-mouth and/or object-to-mouth contacts per hour to be underestimated. A combo-to-mouth category that accounts for both simultaneous types of contacts may provide a more accurate estimate of the indirect ingestion route of exposure.
- An average frequency of 9 hand-to-mouth contacts per hour among 2 to 5 year olds is recommended for regulatory risk assessments (US EPA, 2002). The CPPAES results suggest that a higher value may be appropriate (Table 6.3).

- Figure 6.1 presents the average frequency of hand- and object-to-mouth contacts during all eating and non-eating events. The highest hand-to-mouth frequency was observed in CPPAES.
- Factors affecting hand-to-mouth contact frequencies may include inclusion of eating events, amount of time on tape, types of activities, number of children, and age range.
- An analysis of hand-to-mouth activities in MNCPES has been published by Freeman *et al.* (2001). They reported that hand-to-mouth activities were significantly more frequent (t test, $P<0.05$) among girls than among boys.
- The MNCPES data also showed that hand-to-mouth and object-to-mouth activities were more frequent (Mann–Whitney, $p<0.05$) indoors than outdoors (Freeman *et al.*, 2001).
- Published studies have quantified the hand- and object-to-mouth activities of young children (Zartarian *et al.,* 1998; Reed *et al.,* 1999; Tulve *et al.,* 2002; Freeman *et al.,* 2005). These studies suggest that young children may exhibit higher hand-to-mouth and/or object-to-mouth contacts than older children and adults.
- Standardized approaches for quantifying the activity patterns of children are needed in order to compare results among different studies.

6.2 Residue Loadings on Mouthed Objects and Removal by Mouthing

For indirect ingestion estimates, objects that are commonly mouthed are identified in the field and the residue loadings on these objects are measured. Objects commonly mouthed by preschoolers were identified in CTEPP. Pesticide loadings on toy surfaces were measured in the CHAMACOS and CPPAES studies. Data on the fraction of residues that may be removed by mouthing of fingers was collected in the laboratory-based Transfer studies using non-toxic fluorescent surrogates.

- Objects commonly mouthed by preschoolers were identified in CTEPP. These items were typically toys and food-related items (Table 6.4).
- Chlorpyrifos loadings on toy surfaces were much higher following recent applications, as evidenced by the higher values in CPPAES than in CHAMACOS (Table 6.5). Loading on toy surfaces in CPPAES (Table 6.5) were greater than surface loadings as measured by deposition coupons (Table 4.4).
- Measurements from CPPAES (data not presented) suggest that surface wiping of plush toys yields only a small fraction of the total amount of chlorpyrifos absorbed into the toys (as measured by extraction). Indirect ingestion among children who regularly mouth soft toys may thus be underestimated by toy surface wipes.
- In "transfer off" experiments conducted with a fluorescent tracer (riboflavin) as part of the Transfer studies, removal from skin via the mouthing of 4 fingers was measured. Eight replicates were performed with each of three participants (data not presented), with 0 to 26% of the tracer removed per replicate (loss was significantly different from zero in only one-half of the replicates).

Table 6.1 Collection methods for the transfer of pesticide surface residues to food or objects.

Study	Study Type	Age Range	Sampling Details	Collected After Application	Sample Handling	Composite Sample	Insecticides Measured	Comments
Food (Surfaces to Foods)	Laboratory	n/a	1, 10, & 60 min contact between food and contaminated surfaces	Yes-1 hr following applications	Foods extracted immediately following sampling	No	Chlorpyrifos Diazinon Heptachlor Isofenphos Malathion Permethrin	Surface wipes were collected. The influence from contact force and duration were evaluated
Food (Tile to Foods)	Laboratory	n/a	10 min contact between food and contaminated tile surface	Yes-1 hr following applications	Foods extracted immediately following sampling	No	Chlorpyrifos Cyfluthrin Cypermethrin Deltamethrin Fipronil Malathion Permethrin	Surface wipes and deposition on foil coupons collected
DIYC	Field	1-3 yr	Handled leftover food, untouched leftover food, food press	Yes	Collected in individual zip closure bags	Yes	Diazinon	Foods leftover from meal were combined into two types of samples; *i.e.*, all handled foods combined, all untouched foods combined
CHAMACOS	Field	0.5-2.5 yr	Teething ring or small ball provided 1.5 days before sampling	No	Stored at -20 C until analysis	No	Chlorpyrifos Diazinon Permethrin	Surface of toys wiped
CPPAES	Field	<5 yr	Plush toy given to child to handle for 11 days	Yes	No information	No	Chlorpyrifos	Surface of toys wiped; whole toys extracted
Transfer	Laboratory	Adult	Mouthing removal of fluorescent tracer	n/a	Video-fluorescence imaging	No	Surrogate (Riboflavin)	Many measurements at detection level of technique

n/a, Not applicable

Table 6.2 Videotaped children's hand- and object-to-mouth activity details.

Study	N	Age (years)	Sampling Location	Time Period	Method of Analysis	Activity of Interest	Availability
MNCPES	19	3 to 12	Homes (inside and/or outside)	4 consecutive hours in normal daily activities	Methods of Reed *et al.*, 1999	Hand-to-mouth Object-to-mouth	Freeman *et al.*, 2001.
CPPAES	10	2 to 5	Homes (inside or outside)	4 hours on Day 2 following crack and crevice application of chlorpyrifos	Computer software (Virtual Timing Device) Quantified 4 hours of videotape for both hands	Hand-to-mouth Object-to-mouth	Freeman *et al.*, 2004.

Table 6.3 Videotaped hand-to-mouth and object-to-mouth counts.

Study	Hand-to-Mouth		Object-to-Mouth		Eating Events
	Mean	Median	Mean	Median	
CPPAES (2 to 5 yrs)	19.8	16	8.4	6.4	Unspecified
Tulve [a] ≤24 month old	18	12	45	39	Excluded
Tulve >24 month old	16	9	17	9	Excluded
MNCPES (3 to 12 yrs)	5.7	2.5	1.8	0	Unspecified
MNCPES boys indoor	4.7	NR	1.0	NR	Unspecified
MNCPES girls indoor	8.1	NR	2.6	NR	Unspecified

NR, Not Reported
[a] Tulve data (Tulve *et al.*, 2002) included for comparison.

Table 6.4 Objects commonly mouthed by preschoolers in CTEPP.

Category	Items
Toys	Plastic rings/bracelets, stuffed animals, balls, walkie talkie, building blocks, doll, bubble blower
Food-Related Items	Ice pops, candy wrapper, water bottle, utensils, napkins, drinks
Miscellaneous	Plastic blow-up chair, pens, greeting cards, clothing, CDs, towels, blanket, pets

Table 6.5 Median and 95th percentile pesticide loadings (ng/cm^2) measured on toy surfaces.

Study	Chlorpyrifos		Diazinon		*cis*-Permethrin		*trans*-Permethrin		cyfluthrin	
	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
CHAMACOS	BDL[a]	0.15	0.034	0.27	BDL	0.053	BDL	0.072	BDL	BDL
CPPAES	3.0	21	--[b]	--	--	--	--	--	--	--

[a] BDL, Below minimum detection limit
[b] Blank cells (--) indicate the pesticide was not measured in the study

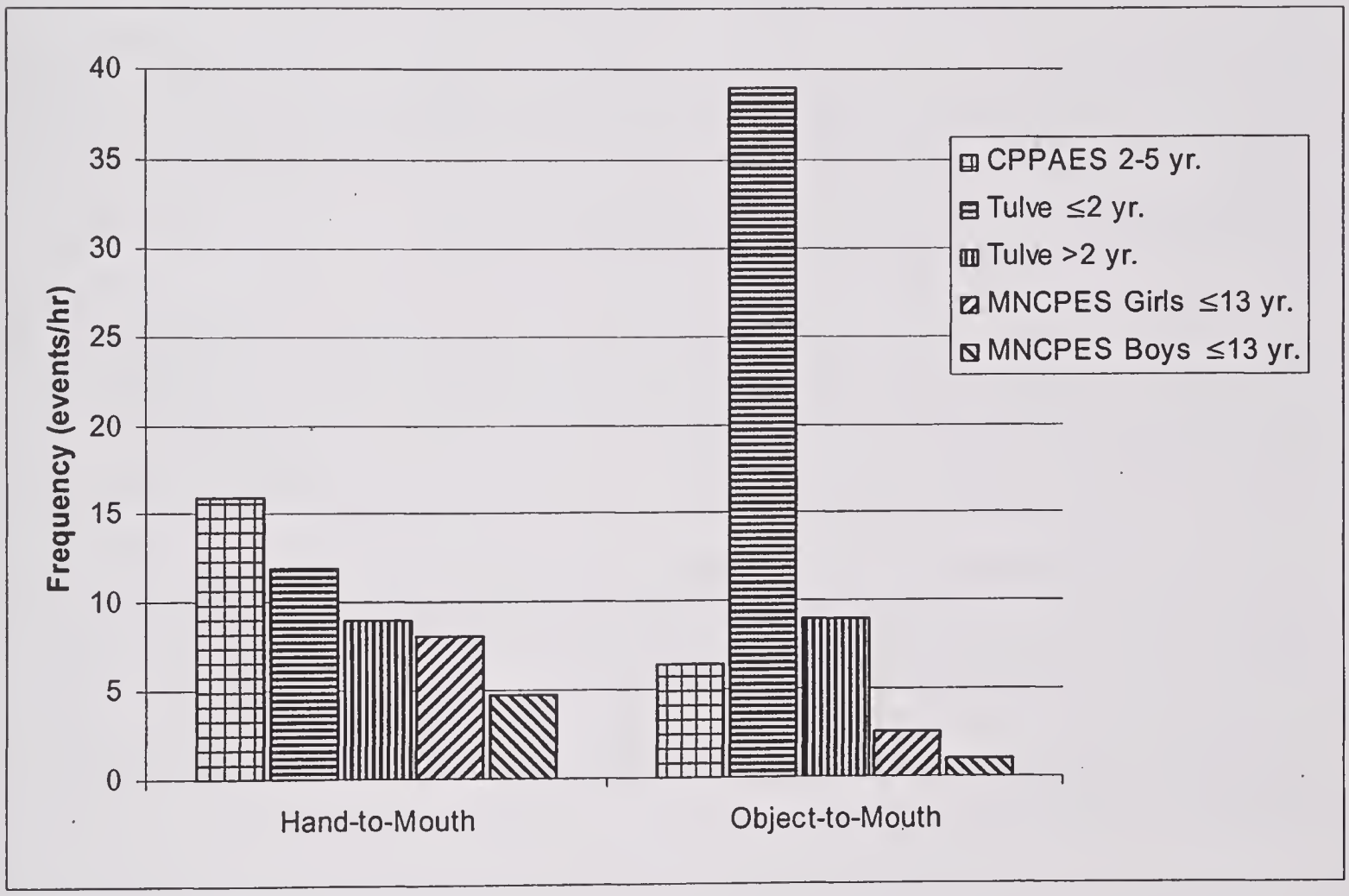


Figure 6.1 Comparison of the median hand-to-mouth and object-to-mouth contacts per hour among CPPAES and MNCPES children. MNCPES values are means instead of medians. Tulve data (Tulve *et al.*, 2002) included for comparison.

6.3 Transfer of Pesticide Residues to Food

- The experiments reported here (Appendix B, Food Transfer Studies) used loadings that were near to or greater than the 95th percentile for loadings in most of the recent field studies (See Table 4.4).
- Higher pesticide transfer to food occurred from hard, smooth surfaces, such as hardwood flooring; lower transfer occurred from carpet. For example, 33% of chlorpyrifos was transferred from wood flooring to an apple, whereas the amount transferred from carpet was not enough to be reliably quantified (Table 6.6).
- Bologna, a moist and fatty food, removed a higher percentage of pesticides from a hard surface than did fruit leather, a low-fat and low-water content food (Table 6.7).
- Comparison (Table 6.8, Figure 6.2) of measured dietary intake of diazinon (incorporating excess contamination due to handling) with estimates predicted by the Children's Dietary Intake Model (CDIM) suggests that use of fixed values for transfer efficiencies and for activity factors in the model may result in inaccurate estimates of daily dietary intake. Model-predicted estimates generally under-predicted intake.
- Diazinon concentrations in untouched leftover food were compared with those in handled leftover food in DIYC. Daily dietary intake estimates accounting for contamination due to handling by children were often double the intake estimates based on untouched food (Total Measured Dietary Intake *vs.* Duplicate Diet Intake, Table 6.8), indicating that duplicate diets may significantly underestimate actual intake in homes that have high surface pesticide residue loadings.
- Food transfer studies have provided evidence that transfer of pesticide residues from surfaces to foods is dependent on such factors as pesticide class, food type, contact duration, and contact force (data not presented).
- Applied force produced a considerable increase in transfer efficiency (data not presented). Moreover, the effect of applied force was even more dramatic as contact duration increased.

Table 6.6 The transfer efficiency (percent transfer, mean ± sd) of pesticide residues from treated surfaces to foods (relative to transfer to IPA wipes), after a 10-min contact duration (Food Transfer Studies).

Pesticide	Sampling Media	N	Treated Surface		
			Ceramic Tile	Wood Flooring	Carpet
Chlorpyrifos (21-38 ng/cm²)	Bologna	2	36 ± 20	15 ± 4	BQL [a]
	Apple	2	18 ± 5	33 ± 8	BQL
	Cheese	2	7 ± 0	26 ± 1	BQL
Diazinon (20-30 ng/cm²)	Bologna	2	41 ± 5	29 ± 0	BQL
	Apple	2	35 ± 8	50 ± 5	BQL
	Cheese	2	20 ± 7	103 ± 18	BQL
Malathion (33-45 ng/cm²)	Bologna	2	60 ± 21	31 ± 1	BQL
	Apple	2	132 ± 74	18 ± 1	212 ± 60
	Cheese	2	94 ± 33	52 ± 37	400 ± 173
cis-Permethrin (40-53 ng/cm²)	Bologna	2	19 ± 15	70 ± 86	BQL
	Apple	2	26 ± 13	3 ± 1	BQL
	Cheese	2	BQL	BQL	BQL
trans-Permethrin (43-55 ng/cm²)	Bologna	2	23 ± 20	10 ± 1	BQL
	Apple	2	29 ± 14	5 ± 0	BQL
	Cheese	2	BQL	BQL	BQL

[a] BQL = Below Quantitation Limit

Table 6.7 The transfer efficiency (percent transfer, mean ± sd) of pesticide residues from a treated ceramic tile surface to various foods and to an IPA Wipe (Food Transfer Studies).

Pesticide Class	Pesticide	Sampling Media	N	% Transfer
Organophosphate	Chlorpyrifos (123 ng/cm²)	Bologna	3	64.7 ± 15.0
		Apple	3	27.5 ± 8.0
		Fruit Leather	3	13.5 ± 2.0
		20-mL IPA Wipe	3	99.8 ± 10.8
	Malathion (193 ng/cm²)	Bologna	3	74.9 ± 17.7
		Apple	3	29.7 ± 8.4
		Fruit Leather	3	8.7 ± 2.7
		20-mL IPA Wipe	3	104.6 ± 10.9
Pyrethroid	Cyfluthrin (143 ng/cm²)	Bologna	3	47.8 ± 13.4
		Apple	3	24.0 ± 3.4
		Fruit Leather	3	0.7 ± 0
		20-mL IPA Wipe	3	108.5 ± 12.1
	Cypermethrin (185 ng/cm²)	Bologna	3	45.0 ± 10.7
		Apple	3	21.5 ± 6.9
		Fruit Leather	3	0.6 ± 0
		20-mL IPA Wipe	3	101.5 ± 7.0
	Deltamethrin (211 ng/cm²)	Bologna	3	39.2 ± 6.1
		Apple	3	22.2 ± 5.1
		Fruit Leather	3	2.4 ± 0.2
		20-mL IPA Wipe	3	83.7 ± 4.3
	Permethrin (147 ng/cm²)	Bologna	3	44.0 ± 11.5
		Apple	3	19.8 ± 7.1
		Fruit Leather	3	1.3 ± 0.1
		20-mL IPA Wipe	3	100.8 ± 4.8
Phenylpyrazole	Fipronil (203 ng/cm²)	Bologna	3	43.3 ± 1.6
		Apple	3	30.9 ± 14.8
		Fruit Leather	3	2.0 ± 1.7
		20-mL IPA Wipe	3	103.8 ± 10.4

Table 6.8 The measured and predicted ingestion (ng/day) of diazinon from the DIYC.

Child	Sampling Day	Duplicate Diet Intake	Excess Dietary Intake [a]	Total Measured Dietary Intake [b]	CDIM Predicted Dietary Intake [c]	Percent Difference [d]
		ng/d	ng/d	ng/d	ng/d	%
1	Pre	197	384	581	357	-39
	1	1063	1270	2333	1271	-46
	4	280	220	500	281	-44
	5	270	501	771	333	-57
	6	140	322	462	142	-69
	7	563	536	1099	702	-36
	8	253	160	413	397	-4
2	1	455	156	611	663	9
	2	233	95	328	402	23
	3	212	373	585	392	-33
	4	260	414	674	612	-9
	5	188	189	377	278	-26
3	2	95	90	185	509	175
	8	412	344	756	940	24

[a] Measured surface-to-food and hand-to-food transfer due to handling of foods, concentration in handled but uneaten portion extrapolated to eaten portion.
[b] Duplicate Diet intake plus Excess Dietary intake.
[c] Estimated by deterministic model using fixed transfer efficiency and activity values.
[d] Percent Difference = 100*[(CDIM Predicted Intake – Total Measured Intake)/(Total Measured Intake)].

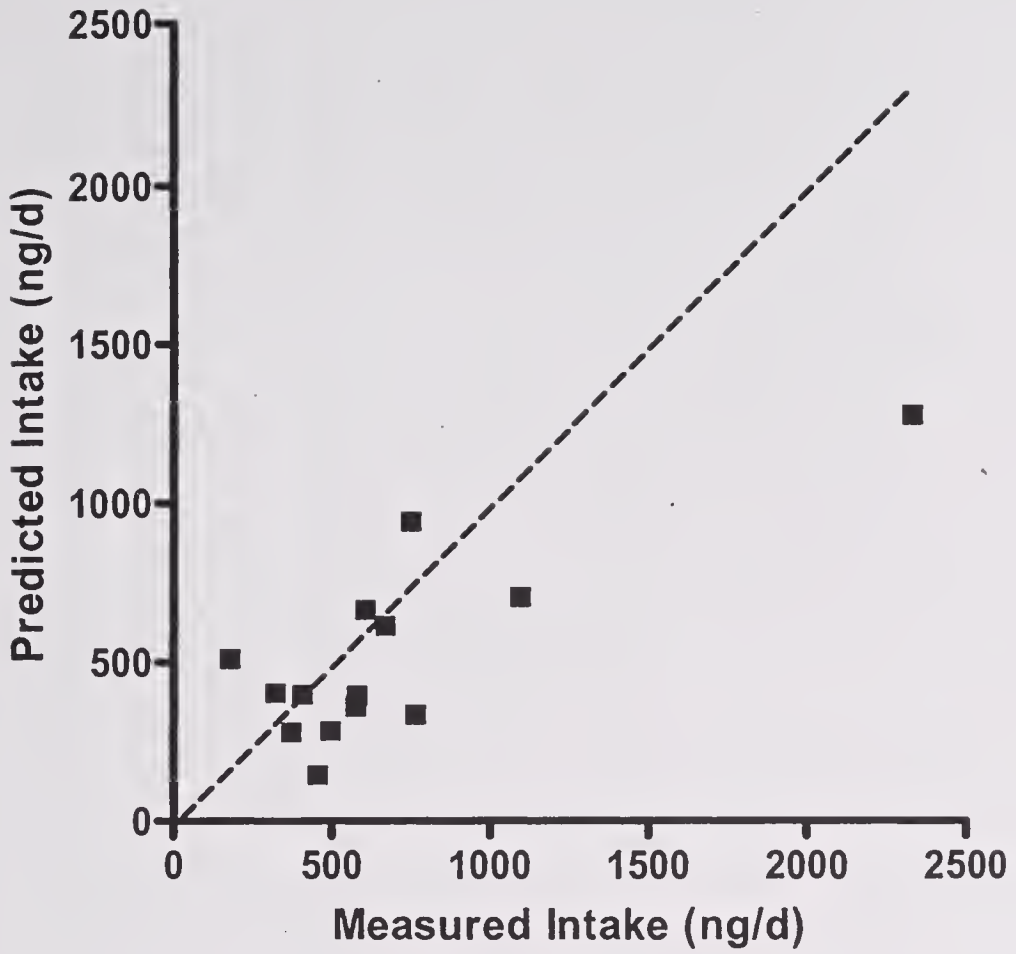


Figure 6.2 Comparison of measured and predicted ingestion of diazinon (ng/day) from the DIYC. Dashed line represents a hypothetical slope of 1. Measured intake generally exceeds predicted intake, as indicated by the majority of points lying to the right of the dashed line.

6.4 Indirect Ingestion of Dust and Soil

The potential indirect ingestion exposure (ng/day) can be estimated using indoor floor dust (ng/g) and outdoor soil sample concentrations (ng/g) together with the child's body weight (kg), estimated daily dust ingestion rate (g/day), estimated daily soil ingestion rate (g/day), and the estimated oral bioavailability. In CTEPP, the daily dust ingestion rates were calculated based on questionnaire responses related to specific activities of each child in the month prior to field sampling. These activities included pacifier use, teething, mouthing body parts, licking floors, and placing toys or other objects into the mouth. The daily soil ingestion rates were estimated based on how often a child played with sand/dirt and ate dirt, sand, or snow. Many of these parameters have very high uncertainty associated with them. The daily dust and soil ingestion rates were each estimated as 0.025, 0.050, or 0.100 g/day. The indirect exposure estimates, presented in Table 6.9, showed the following:

- Indirect ingestion estimates for the permethrin isomers were much higher than for chlorpyrifos or diazinon, largely because permethrin was measured at much higher concentrations in floor dust (Figures 4.6 and 4.7).
- The differences between NC and OH in mean permethrin concentrations in dust suggest potential regional differences in indirect ingestion.

Table 6.9 The estimated exposures (ng/day) of NC and OH preschool children in the CTEPP study to chlorpyrifos, diazinon, and permethrin through indirect ingestion.

Pesticide	State	N	Mean	SD	GM	GSD	Min	P25	P50	P75	P95	Max
Chlorpyrifos	NC	117	15.5	29.0	6.2	1.3	0.3	2.8	5.2	14.8	80.4	233
	OH	116	27.8	164	3.0	1.5	0.2	1.1	2.7	6.2	33.5	1570
Diazinon	NC	118	21.7	81.9	1.6	2.0	<MDL	0.4	1.0	4.3	150	622
	OH	116	49.1	367	1.5	1.9	<MDL	0.4	1.0	3.4	45.3	3800
cis-Permethrin	NC	120	220	670	48.4	1.6	1.7	17.1	48.1	113	718	4540
	OH	116	61.5	139	21.3	1.4	1.9	7.8	17.9	52.7	327	1210
trans-Permethrin	NC	120	222	698	42.7	1.7	1.1	11.9	35.4	119	680	4800
	OH	102	61.2	153	16.6	1.5	1.2	5.3	11.7	45.9	210	1190

<MDL, less than method detection limit

6.5 Indirect Ingestion: Summary

As shown in the bulleted lists of observations from these laboratory and observational studies, progress has been achieved in identifying and quantifying a number of factors that are believed to potentially impact indirect ingestion among children.

- Videotape analysis of children's hand- and object-to-mouth contacts has provided evidence that hand-to-mouth activities were more frequent: among infants and toddlers than among older children, among girls than among boys, and at indoor locations than at outdoor locations.
- Objects most commonly mouthed by preschoolers were identified as typically being toys and food-related items.
- High chlorpyrifos loadings were measured on toy surfaces following routine residential application.
- Fluorescent tracer experiments found that removal from skin (at very high tracer loadings) by mouthing was highly variable. Additional information is still needed on the fraction of residue transferred from the hands to mouth during typical mouthing events at dermal loading levels observed in field studies.
- At high surface loadings, pesticide transfer to food was greater from hard, smooth surfaces than from carpet.
- In homes with high surface pesticide residue loadings, residue concentrations in foods handled by children were often twice as high as concentrations in leftover unhandled foods.
- The transfer of pesticide residues from surfaces to foods appears to be dependent on such factors as pesticide class, food type, contact duration, and applied force.
- Indirect ingestion estimates for permethrin were much higher than for chlorpyrifos or diazinon, largely because permethrin was measured at much higher concentrations in floor dust.

7.0 DERMAL EXPOSURE MEASUREMENTS

The ability to accurately estimate surface-to-skin transfer of contaminants from intermittent contacts remains a critical and missing link in pesticide exposure and risk assessments. For children's exposures, transfer of chemicals from contaminated surfaces such as floors and furniture is potentially significant. Once on the skin, residues and contaminated particles can be transferred back to the contaminated surface during subsequent contact, lost by dislodgement or washing, or transferred into the body by percutaneous absorption or hand-to-mouth activity. A better understanding of the relevant factors influencing transfers from contaminated surfaces to skin and the resulting dermal loading will reduce uncertainty in exposure assessment. Areas of uncertainty with respect to dermal transfer are related to the important factors that impact transfer, whether or not a steady-state condition is reached, and the conditions that affect removal. Laboratory tests were conducted by NERL using nontoxic fluorescent tracers to evaluate significant transfer parameters. The results of these tests are described in this section (Section 7.1).

Measurements of pesticide residues on children's hands have been performed in a number of studies. Both hand wipe and hand rinse methods have been used. The collection efficiency of different wipe and rinse methods can be expected to differ, with an eight-fold difference reported between hand rinses and hand wipes in one study (Hore, 2003). Furthermore, differences in dermal exposure and dose due to free pesticide residue versus particle- (or dust-) bound pesticides may be important in interpreting the results. Results of wipes and rinses in selected studies are summarized in tables and figures presented below (in Section 7.2).

An alternative approach for estimating dermal exposure is the cotton garment surrogate. Similar to the approach used for measuring occupational exposures to pesticides, cotton garments, which can consist of a bodysuit and/or socks, have been used in three studies that are reported below (Section 7.3).

Important Factors Affecting Transfer

Dermal exposure to surface residues is dependent on human activities that result in contact with surfaces and the physicochemical and mechanical mechanisms of transfer of residues from the surface to the skin. Several factors are commonly believed to affect transfer (Table 7.1). These factors can be grouped as characteristics of the surface (including contaminant loading, type of surface, and temperature), of the contaminant (including formulation, physical state, particle size, vapor pressure, viscosity, water solubility, lipophilicity, and being particle-bound), of the skin (including moistness and contact area), of contact (including duration, force, frequency, motion, and interval), and of protection measures (including clothing and hand washing).

Many of these have previously been investigated, though not necessarily specific to pesticides and skin. Kissel *et al.* (1996) reported *moisture content* and *particle sizes* of soil to be significant factors affecting the process of adherence to skin. Rodes *et al.* (2001) reported that only about 1/3 of the palm contacted surfaces during a press and that dust-to-skin transfer increased with hand dampness, decreased as surface roughness increased, and decreased with consecutive presses (requiring about 100 presses to reach equilibrium). Brouwer *et al.* (1999) reported that

whereas only 4-16% of the surface area of the palm of the hand is covered with a fluorescent tracer after one contact with a hard surface, about 40% becomes covered after twelve consecutive contacts. At least three studies have investigated the transfer of pesticides from surfaces to hands (measured using IPA wipes of hands.). Briefly, Lu and Fenske (1999) reported transfer of chlorpyrifos residues to hands to be 0.04 to 0.26% from carpets and 0.69% from furniture. Camann *et al.* (1996) examined transfer from nylon carpet to dry or moistened hands and reported transfers ranging from 0.7–1.3% for chlorpyrifos, 2.9–4.8% for pyrethrin I, and 1.5–2.8% for piperonyl butoxide. Clothier (2000) examined transfer of the same residues from vinyl sheet flooring and reported transfers of 1.5% to dry and 4.4-5.2% to wet skin for chlorpyrifos, 3.6% (dry) and 8.9 – 11.9% (wet) for pyrethrin I, and 1.4% (dry) and 4.1-4.8% (wet) for piperonyl butoxide.

7.1 Laboratory Fluorescent Measurement Studies

Laboratory tests were performed to evaluate transfer efficiencies (TEs) of nontoxic fluorescent tracers (as surrogates for pesticide residues) from common household surfaces to hands (Cohen Hubal *et al.*, 2005). The laboratory studies evaluated parameters affecting surface-to-hand transfer, including surface type, surface loading, contact motion, pressure, duration, and skin condition in two sets of experiments (Table 7.2). The data from the laboratory fluorescent measurement studies are presented in Tables 7.3 to 7.6 and Figures 7.1 and 7.2.

- Tests comparing fluorescent tracers with pesticides (Figure 7.1) showed that the transfer of riboflavin to PUF rollers and C18 disks is similar to that of chlorpyrifos, and that the transfer of Uvitex is similar to that of the pyrethroids permethrin and esfenvalerate.
- Laboratory studies using fluorescent tracers riboflavin and Uvitex OB (Tables 7.3 to 7.6) indicated that *tracer type*, *surface type*, *contact motion*, and *skin condition* were all significant factors. Transfer was greater with laminate (over carpet), smudge (over press), and sticky skin (over moist or dry). *Contact duration* and *pressure* (force) were not important factors.
- Comparison of "first contact" to "repeated contact" results (Table 7.4) suggests that the effect of surface type appears to diminish with repeated contact while the effect of skin condition (moist vs. dry) appears to increase with repeated contact.
- Laboratory surface loadings (0.2 and 2.0 $\mu g/cm^2$) were much higher than the median values of 0.032 and 0.0014 $\mu g/cm^2$ measured by deposition coupons (Table 4.4) after crack and crevice application of chlorpyrifos in the Test House and CPPAES studies, respectively,
- In the initial tracer experiments with high surface loadings, dermal loadings appear to reach a maximum by the fourth or fifth contact (data not presented), suggesting a saturation effect. In the follow-up experiments with lower surface loadings (Figure 7.2), dermal loadings appear to increase linearly through the seventh contact, suggesting that at lower surface loadings, more contacts may be required to reach steady state.
- In "transfer off" experiments described earlier (Section 6.2), the amount removed from fingers by mouthing was significantly different from zero in only half of the replicates.

Table 7.1 Factors commonly believed to affect dermal transfer.

Category	Parameter	Source
Surface	Level of contamination	Goede *et al.*, 2003; This Report
	Type of surface: roughness, carpet vs. hard surface	Brouwer *et al.*, 1999; Rodes *et al.*, 2001
Contaminant	Formulation	Marquart *et al.*, 2005
	Physical state: solid, liquid	Marquart *et al.*, 2005
	Particle characteristics: particle size distribution, moistness	Kissel *et al.*, 1996
	Liquid characteristics: viscosity and related properties	Marquart *et al.*, 2005
	Physical properties of active ingredient: vapor pressure, water solubility, lipophilicity	This Report
Skin	Moistness	Camann *et al.*, 1996; Clothier, 2000; Rodes *et al.*, 2001; This Report
	Contact area	Brouwer *et al.*, 1999
Contact	Frequency: number of contacts or objects	Brouwer *et al.*, 1999; Rodes *et al.*, 2001; This Report
	Interval between contacts	Camann *et al.*, 1996;
	Motion: press, smudge, drag	Lu and Fenske, 1999;
Protection	Clothing: use, area covered, material	Marquart *et al.*, 2005
	Hand washing: frequency	This Report

Categories and parameters modified from Marquart *et al.*, 2005.

Table 7.2 Study parameters tested in surface-to-skin transfer experiments in the Characterizing Pesticide Residue Transfer Efficiencies study.

Parameter	Initial Experiments	Refined Experiments [a]
Tracer	Riboflavin [a]	Riboflavin [b] or Uvitex [c]
Skin Condition	Dry, Moist, or Sticky	Dry or Moist
Surface Type	Carpet or Laminate	Carpet or Laminate
Surface Loading	2 or 10 $\mu g/cm^2$	0.2 or 2 $\mu g/cm^2$
Contact Motion	Press or Smudge	Press or Smudge
Contact Duration	2 sec or 20 sec	--[d]
Contact Pressure	7 or 70 kg/cm^2	--
Contact Number	Multiple	Multiple

[a] Refined experiments added Uvitex, reduced the loading levels, and reduced the number of parameters tested
[b] Relatively water soluble
[c] Relatively water insoluble
[d] Blank cells indicate that parameter was not investigated in the study

Table 7.3 Skin loadings (mean, standard deviation) measured following surface-to-skin transfer experiments (initial experiments). (Source: Cohen Hubal *et al.*, 2005.)

Contact	Hand condition			Surface type		Surface loading	
	Dry	Moist	Sticky	Carpet	Laminate	High	Low
Skin loading, $\mu g/cm^2$, average (SD) [a]							
1	0.3 (0.6)	0.4 (0.3)	0.7 (0.6)	0.4 (0.5)	0.5 (0.5)	0.6 (0.6)	0.3 (0.3)
2	0.4 (0.4)	0.9 (0.6)	1.2 (0.7)	0.8 (0.7)	0.8 (0.6)	1.0 (0.7)	0.6 (0.5)
3	0.5 (0.5)	1.0 (0.6)	1.5 (0.7)	1.0 (0.8)	1.0 (0.7)	1.2 (0.8)	0.8 (0.6)
4	0.6 (0.5)	1.3 (0.8)	1.6 (0.8)	1.2 (0.9)	1.2 (0.7)	1.4 (0.8)	0.9 (0.7)
5	0.5 (0.3)	1.3 (0.7)	1.8 (0.8)	1.3 (1.0)	1.0 (0.6)	1.4 (0.9)	0.9 (0.7)
Skin loading, $\mu g/cm^2$ (without sticky hand condition), average (SD)							
1	0.3 (0.6)	0.4 (0.3)		0.4 (0.6)	0.3 (0.2)	0.5 (0.6)	0.2 (0.2)
2	0.4 (0.4)	0.9 (0.6)		0.7 (0.7)	0.6 (0.4)	0.8 (0.7)	0.4 (0.3)
3	0.5 (0.5)	1.1 (0.6)		0.8 (0.8)	0.8 (0.5)	1.0 (0.7)	0.5 (0.4)
4	0.6 (0.5)	1.3 (0.8)		1.0 (0.9)	0.9 (0.5)	1.2 (0.8)	0.6 (0.4)
5	0.5 (0.3)	1.3 (0.7)		0.9 (0.9)	0.8 (0.4)	1.1 (0.8)	0.6 (0.4)

[a] Three subjects provided three independent replicates for each experiment

Table 7.4 Statistical analysis results (p-values) from initial surface-to-hand transfer experiments (Riboflavin).

Analysis	Tracer	Surface Type	Surface Loading	Contact Motion	Pressure	Duration	Skin Condition	Contact Number
First contact (ANOVA)								
Transfer efficiency (%)	----------	p<0.1	**p<0.001** [a]	**p<0.05**	p>0.1	p>0.1	**p<0.001**	----------
Loading (ug/cm²)	----------	p>0.1	**p<0.05**	**p<0.05**	p>0.1	p>0.1	**p<0.05**	----------
First contact, sticky hand excluded (ANOVA)								
Transfer efficiency (%)	----------	p>0.1	**p<0.001**	p>0.1	p>0.1	p>0.1	**p< 0.001**	----------
Loading (ug/cm²)	----------	p>0.1	p<0.1	**p<0.05**	p>0.1	p>0.1	p>0.1	----------
Repeated contact (Mixed-Effects Model)								
Loading (ug/cm²)	----------	p>0.1	**p<0.001**	**p<0.001**	p>0.1	p>0.1	**p<0.001**	**p<0.001**
Repeated contact, sticky hand excluded (Mixed-Effects Model)								
Loading (ug/cm²)	----------	p>0.1	**p<0.001**	**p<0.01**	p>0.1	p<0.1	**p<0.001**	**p<0.001**

[a] **Bold text** indicates the parameter is significant.

Table 7.5 Statistical analysis results (p-values) from refined, follow-up surface-to-hand transfer experiments (Riboflavin and Uvitex).

Analysis	Tracer	Surface Type	Surface Loading	Contact Motion	Pressure [a]	Duration [a]	Skin Condition	Contact Number
First Contact (ANOVA)								
Transfer efficiency (%)	**p<0.05** [b]	**p<0.05**	**p<0.01**	p<0.1	----------	----------	p>0.1	----------
Loading (μg/cm²)	p=0.1	**p<0.05**	**p=0.001**	**p<0.001**	----------	----------	p>0.1	----------
Repeated Contact (Mixed-Effects Model)								
Loading (μg/cm²)	**p<0.01**	p=0.1	**p<0.001**	**p<0.001**	----------	----------	**p<0.05**	**p<0.001**

[a] Pressure and duration not included in the follow-up experiments.
[b] **Bold text** indicates the parameter is statistically significant at p<0.05.

Table 7.6 Evidence of importance of factors tested across surface-to-skin transfer experiments.

Parameter	Initial Experiments	Refined Experiments
Tracer	--	●○
Skin Condition	●○	●○
Surface Type	○○	●○
Surface Loading	●○	●●
Contact Motion	●●	●○
Contact Duration	○○	--
Contact Pressure	○○	--
Contact Number	●●	●●

-- not tested
○○ not found to be significant
●○ mixed results or marginally significant at $p<0.10$
●● significant at $p<0.05$ in all tests

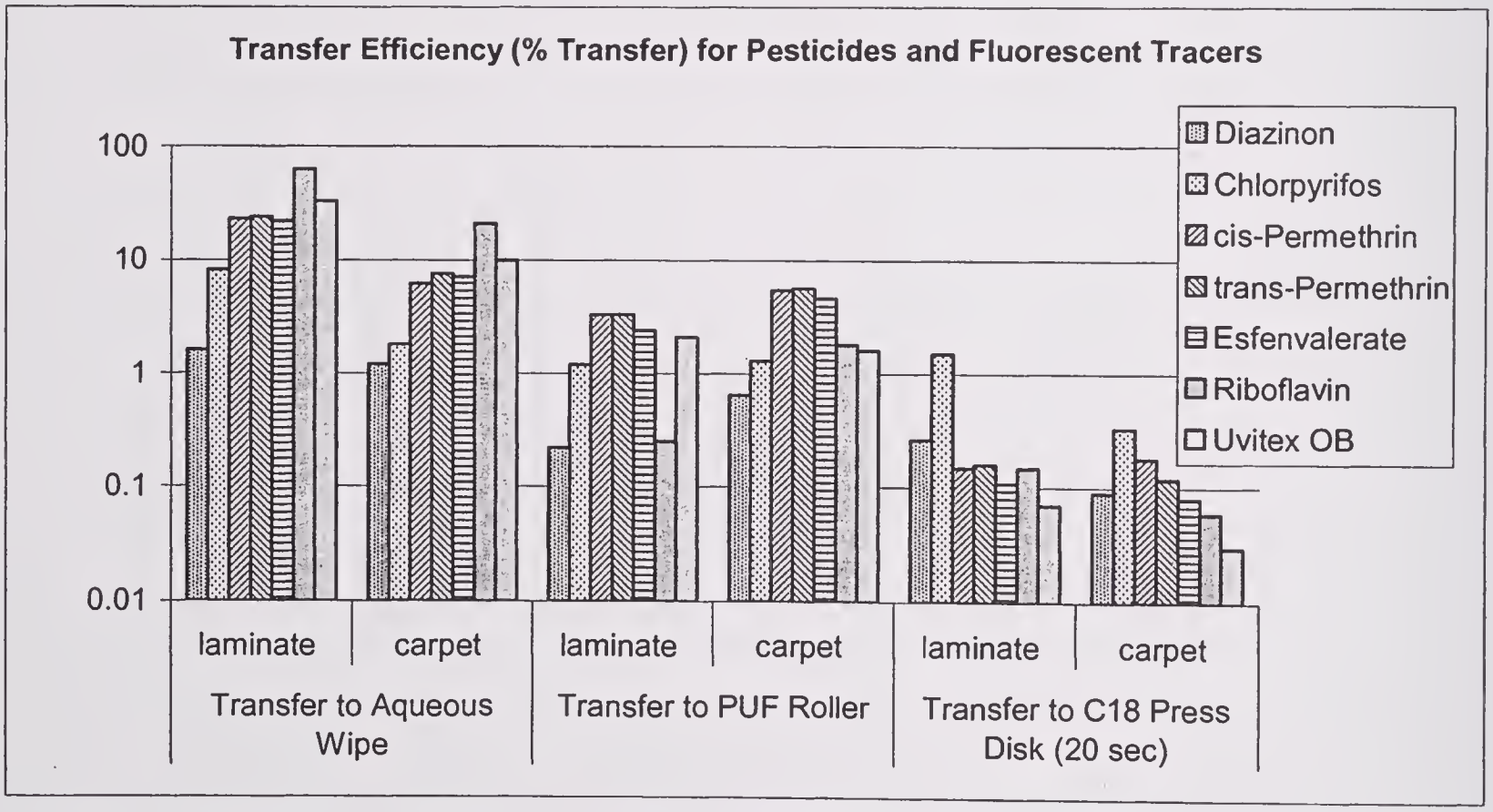
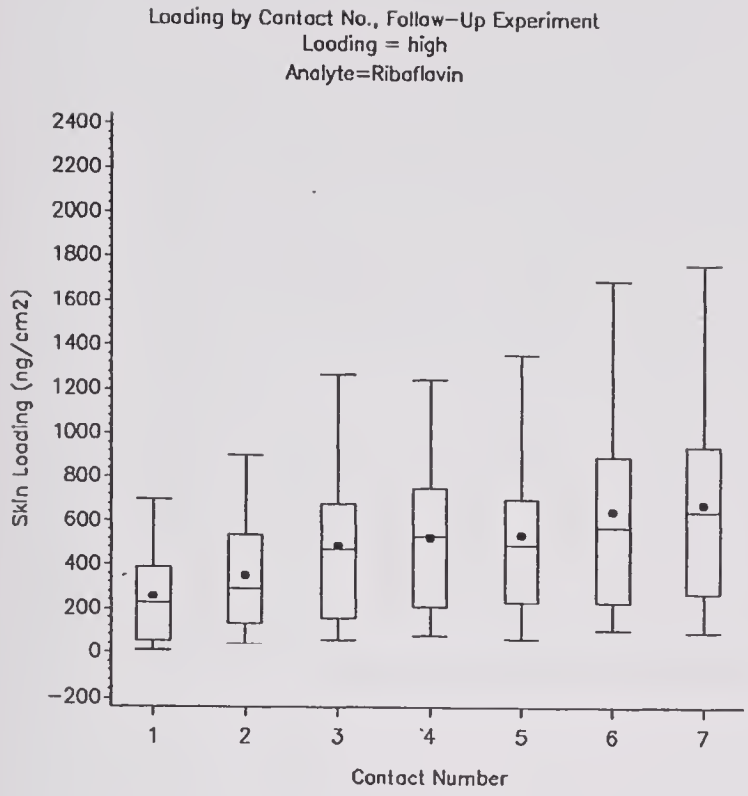
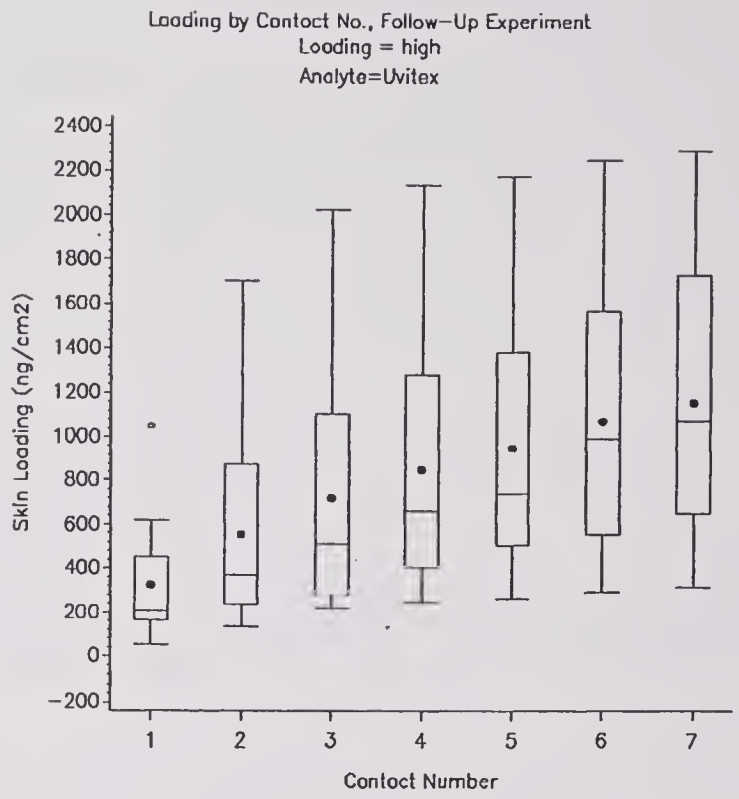
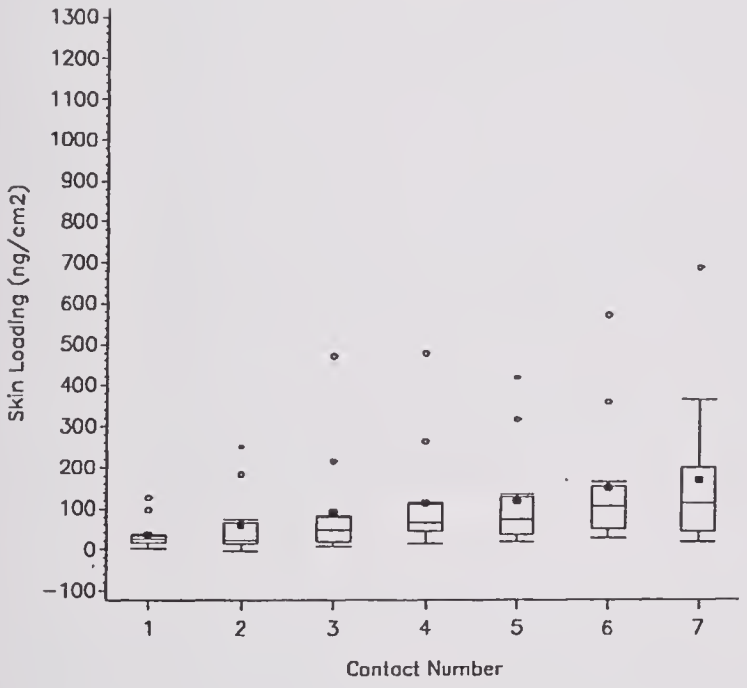


Figure 7.1 Comparison of transfer efficiencies of fluorescent tracers and pesticides from laminate and carpet surfaces to various sampling media.







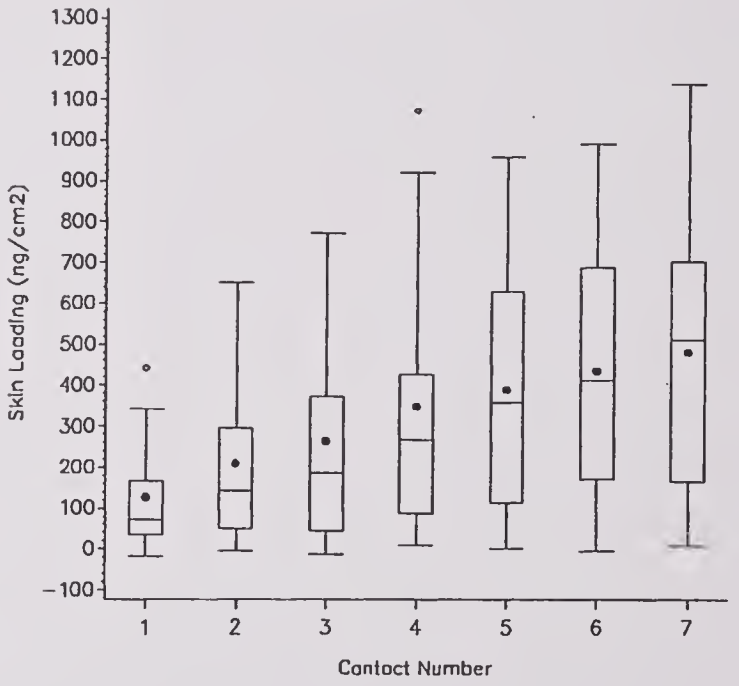


Figure 7.2 Hand loading by contact number, from the refined, follow-up experiments using Riboflavin (left panels) or Uvitex (right panels) with 2 $\mu g/cm^2$ (high) (top panels) or 0.2 $\mu g/cm^2$ (low) (bottom panels) surface loadings. In these particular box-and-whisker plots, means and outliers (below 5th or above 95th percentiles) are represented by dots.

7.2 Measurements of Pesticides on Hands by Wipe and Rinse Methods

Measurements of pesticide residues on children's hands have been performed in the MNCPES, CTEPP, CPPAES, PET, and DIYC studies. Collection efficiencies may vary among studies for a number of reasons. The method of wiping the surfaces of the hand may vary when performed by different researchers or by study participants themselves. Hand rinses may be more effective than hand wipes. Whether the method is a hand wipe or hand rinse, collection efficiency may differ for free pesticide residues versus particle-bound residue. Most of the data presented in this section were collected with hand wipes, except for MNCPES, in which rinses were collected. Both hand wipes and rinses were collected in CPPAES (with mean hand rinse to hand wipe ratios ranging from 4.1 to 7.8 by home). The amount of isopropanol used to collect the hand wipes/ rinses varied by study. A major issue associated with interpreting results of these measurements is the amount of a pesticide on the surface of skin that is never absorbed into the bloodstream. Solvents may extract some of pesticide from top layers of skin, though the extent of extraction will be a function of many factors including pesticide properties.

Methods

In CTEPP, hand wipe samples were collected from 257 preschool children using cotton sponges (SOF-WICK gauze pad; 4" x 4" – 3 ply; Johnson & Johnson) that were pre-cleaned and wetted with 2 mL of 75% isopropanol. The adult caregiver wiped the front and back of both hands of the child. A total of four wipe samples were collected over a 48-hr period (two per day, one before lunch and dinner, before washing hands). Samples were composited (combined) before analysis. The MNCPES hand rinses were collected at home from 102 children on day 1 of the 7-day monitoring period. A technician placed each of the child's hands into a separate zip-closure bag containing 150 mL of isopropanol. Each hand's sample was analyzed separately. The feasibility portion of the PET study collected hand wipes on multiple days from two children after a granular application of diazinon to the lawn by the homeowner. The cotton sponges (SOF-WICK gauze pad; 4" x 4" – 6 ply; Johnson & Johnson) were presoaked with 20 mL of isopropanol. Each child wiped the front and back of each hand. A total of five samples were collected from each child and each was analyzed separately. The CPPAES hand wipe samples were collected from 10 children on multiple days following a professional crack and crevice application of chlorpyrifos. Separate swabs that were wetted with an unreported amount of isopropanol were used to wipe the front and back of each hand. A small number of hand rinse samples were also collected. The DIYC study collected hand wipes on multiple days from three children after a crack and crevice application of diazinon. Each of two gauze pads, pre-wetted with 10 mL of isopropanol, was used to wipe both hands. The two wipes were extracted and analyzed as one sample. In all studies, the surface area of the children's hands was measured.

Results

Table 7.7 summarizes the detection limits for the studies. The median and 95th percentile concentrations are presented in Table 7.8. Individual hand loading measurements are presented in Tables 7.9. Relationships among populations and locations are illustrated in Figures 7.3 to 7.9 and highlighted below.

- In the large observational field studies (Figure 7.3, Table 7.8), the loadings of chlorpyrifos on children's hands measured with rinses in MNCPES were higher than the loadings measured with wipes in the other studies.
- For all compounds, the hand loadings measured with hand wipes in the large observational field studies did not differ substantially (Figure 7.3, Table 7.8).
- Median chlorpyrifos loadings on children's hands (Figure 7.4) were much higher in CPPAES, where homes had recent crack and crevice applications, than in the large observational CTEPP and MNCPES studies.
- Median diazinon loadings on children's hands in the small, pilot-scale PET (lawn application) and DIYC (crack and crevice application) studies were much higher than in the large observational field study CTEPP (Figure 7.4).
- Comparison of hand rinse and hand wipe samples collected from the same participants in CPPAES suggests that hand rinses were more effective at removing residues (Table 7.9).
- Hand rinses may be more efficient than hand wipes at removing chlorpyrifos from the skin, but no information is available on which method better reflects the amount of pesticide that is either absorbed (dermal absorption) or potentially transferred to the mouth (indirect ingestion).
- In the CTEPP study, the median chlorpyrifos hand loadings were higher in NC than OH (at both homes and daycares), suggesting greater chlorpyrifos usage in NC than in OH. Permethrin levels were only slightly higher in NC than in OH (Figure 7.4).
- At residential levels observed in CTEPP, median hand wipe-to-surface loading ratios reach or exceed 1 for the pesticides of interest (Figure 7.5). Please note that floor wipe loadings were measured using an IPA wipe method that was not as efficient as typical wipe methods (Section 4.4).
- A strong relationship is evident in Figure 7.6 between CTEPP hand loadings measured at homes and those measured at daycares for chlorpyrifos (R^2=0.47), diazinon (R^2=0.44), and permethrin (R^2=0.41). The relationship is weak for the degradation product TCPy (R^2=0.03).
- There was a strong relationship between children's hand wipe loadings and adult hand wipe loadings for chlorpyrifos (R^2=0.64; β=0.77), diazinon (R^2=0.77; β=0.81), and permethrin (R^2=0.49; β=0.65) measured in CTEPP (Figure 7.7), despite largely different activity patterns between children and adults.
- Based on regressions of CTEPP hand wipe measurements on either floor dust or floor wipe measurements for chlorpyrifos, diazinon, and permethrin (Figures 7.8 and 7.9), better relationships were observed between hand wipe and floor dust measurements (Figure 7.9) than between hand wipe and floor wipe measurements (Figure 7.8).

Table 7.7 Limits of detection (ng/cm^2) for dermal measurements by compound and study.

Study	Sample type	Chlorpyrifos	Diazinon	*c*-Permethrin	*t*-Permethrin
NHEXAS-AZ	Hand wipe	0.004	0.016	--[a]	--
MNCPES	Hand rinse	0.06	0.08	--	--
CTEPP	Hand wipe	0.003	0.003	0.003	0.003
CPPAES	Hand wipe	NA[b]	--	--	--
CPPAES	Hand rinse	NA[b]	--	--	--
DIYC	Hand wipe	--	0.02	--	--
PET	Hand wipe	--	0.01	--	--

[a] Blank cells indicate that the pesticide was not measured in the study.
[b] Detection limit information unavailable.

Table 7.8 Median and 95th percentile values of pesticide hand loadings (ng/cm^2) measured by hand rinse (HR) or hand wipe (HW) in the large observational field studies.

		Chlorpyrifos		Diazinon		*c*-Permethrin		*t*-Permethrin		Cyfluthrin		TCPY		IMP	
Study	Type	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
NHEXAS-AZ	HW	0.01	0.1	0.015	0.1	--[a]	--	--	--	--	--	--	--	--	--
MNCPES	HR	0.07	0.3	0.07	0.1	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h[b]	HW	0.02	0.3	0.003	0.1	0.1	1.5	0.1	1.3	0.03	0.4	0.02	0.1	--	--
CTEPP-NC d	HW	0.02	0.1	0.01	0.1	0.1	0.3	0.04	0.3	0.03	0.3	0.01	0.03	--	--
CTEPP-OH h	HW	0.01	0.2	0.003	0.1	0.03	0.8	0.03	0.8	0.03	0.1	0.01	0.03	0.003	0.02
CTEPP-OH d	HW	0.01	0.1	0.003	0.04	0.04	0.6	0.03	0.8	--	--	0.01	0.03	0.003	0.02

[a] Blank cells indicate that the pesticide was not measured in the study.
[b] CTEPP: h = home, d = daycare

Table 7.9 Comparison of chlorpyrifos and diazinon loadings (ng/cm²) on children's hands measured with hand rinse (HR) and hand wipe (HW) methods.

Study	Participant	Pre-Appl [a]		Day 1		Day 3		Day 5		Day 7		Day 9		Day 11		3rd Week	
		HR	HW	HR	HW	HR	HW	HR	HW	HR	HW	HR	HW	HR	HW	HR	HW
CPPAES (chlorpyrifos)	Child 1 (4 yr)	-- [b]	--	--	--	--	--	--	--	0.7	--	--	--	--	--	--	--
	Child 2 (4 yr)	0.53	--	5.2	--	18	--	1.6	--	3.8	--	2.3	--	2.3	--	--	--
	Child 3 (4 yr)	--	--	11	--	2.3	--	--	--	3.8	--	2.6	--	2.6	--	--	--
	Child 4 (2 yr)	0.57	--	--	0.79	--	0.34	--	0.81	1.3	--	--	--	--	0.32	--	21
	Child 5 (4 yr)	0.09	--	--	0.3	--	1.4	--	0.28	--	0.37	1.3	--	--	--	--	0.04
	Child 6 (3 yr)	--	0.57	--	0.36	--	0.67	--	0.35	--	0.68	2.3	--	--	0.08	--	0.5
	Child 7 (3 yr)	2.3	--	--	0.17	--	0.25	--	0.22	--	0.51	--	--	--	0.39	--	0.44
	Child 8 (3 yr)	0.21	--	--	0.1	--	0.01	--	0.02	--	0.02	0.26	--	--	0.02	--	0.02
	Child 9 (4 yr)	--	0.07	--	0.08	--	--	--	0.09	--	--	0.74	--	--	0.05	--	0.09
	Child 10 (4 yr)	--	0.43	--	0.43	--	0.68	--	0.5	--	0.36	1.8	--	--	0.27	--	0.41
PET (diazinon)	Child 1 (6 yr)	--	0.01	--	0.6 [c]	--	0.9	--		--		--	0.2	--	--	--	0.2
	Child 2 (10 yr)	--	0.7	--	0.7 [c]	--	0.6	--		--		--	0.1	--	--	--	0.2
DIYC (diazinon)	Child 1 (2 yr)	--	0.06 [d]	--	--	--	--	--	0.14 [de] 0.08 [ef]	--	0.13 [g] 0.21 [fg]	--	0.19 [h] 0.20 [h]	--	--	--	--
	Child 2 (3 yr)	--	--	--	--	--	0.03	--	<MDL [f]	--	--	--	--	--	--	--	--
	Child 3 (1 yr)	--	--	--	--	--	0.10 0.10 [b]	--	0.11	--	0.13	--	--	--	--	--	--

[a] Pre-Appl, Pre-application; [b] Blank cells (--) indicate no measurement; [c] Day 0; [d] Collected from only the right hand of the child; [e] Day 4; [f] Two hand wipe samples were collected on that day: one before breakfast and the other one before supper or bedtime; [g] Day 6; [h] Day 8; [f] <MDL, less than method detection limit.

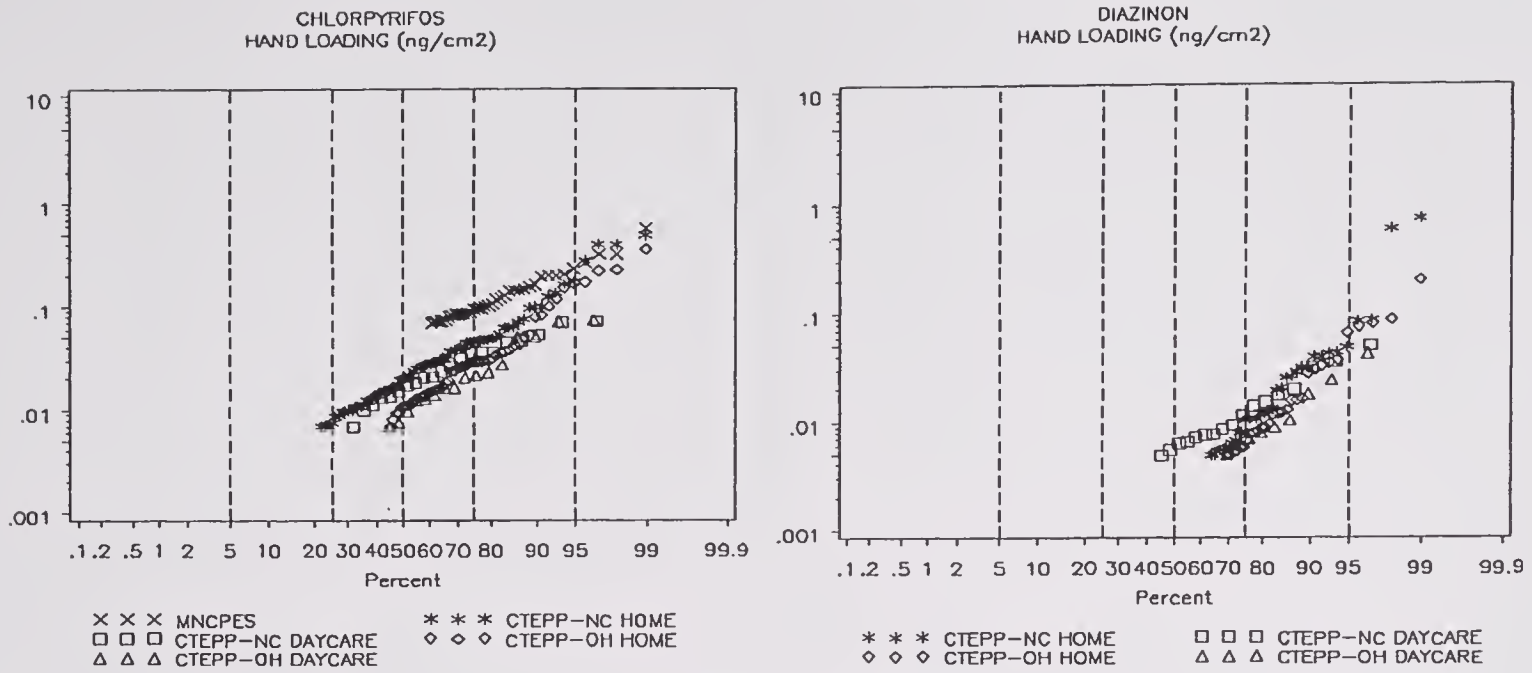


Figure 7.3 Log probability plots of hand loadings (MNCPES data are hand rinses, all others are hand wipes).

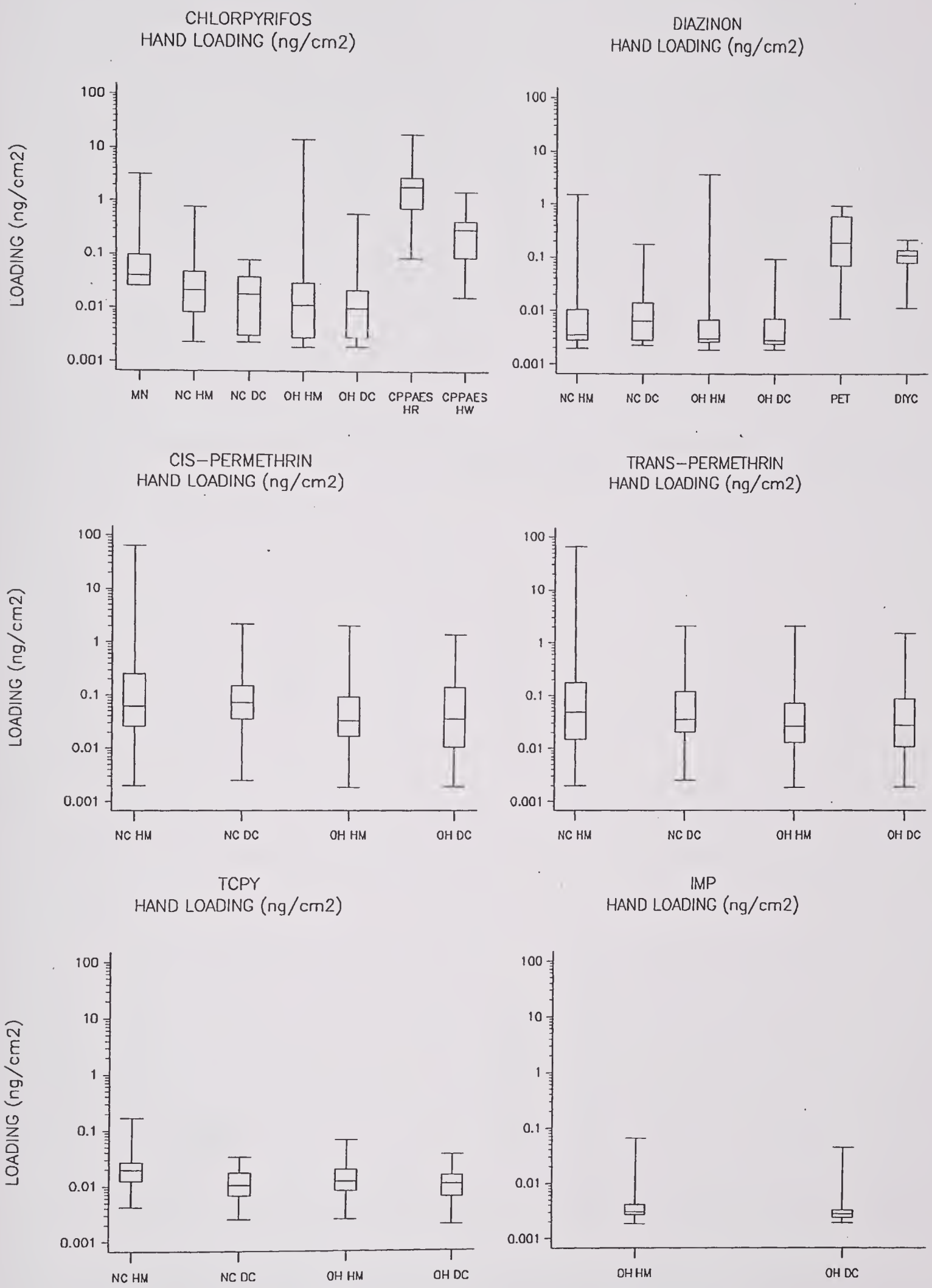


Figure 7.4 Comparison of hand loadings across studies. MNCPES data are hand rinses, CPPAES includes both hand rinses (HR) and hand wipes (HW), all others are hand wipes.

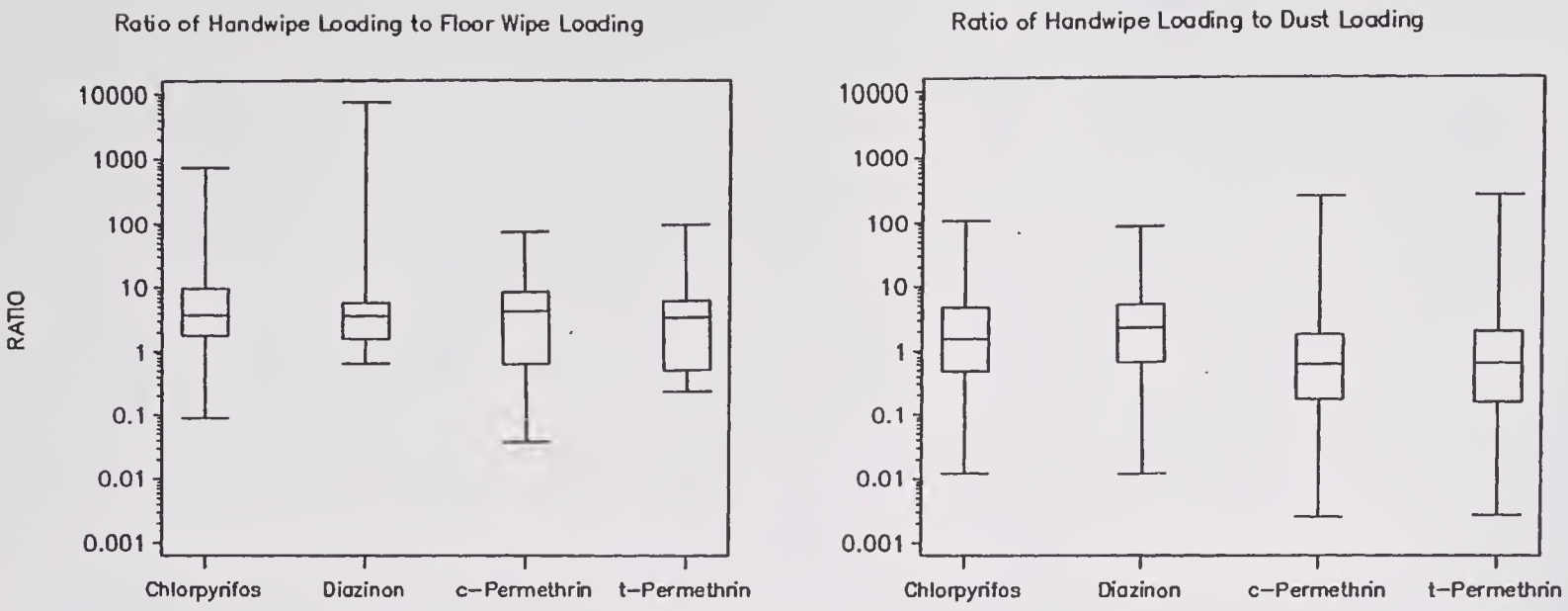


Figure 7.5 Ratios of hand wipe loading to floor wipe loading (left panel) and hand wipe loading to dust loading (right panel) for pesticides in CTEPP.

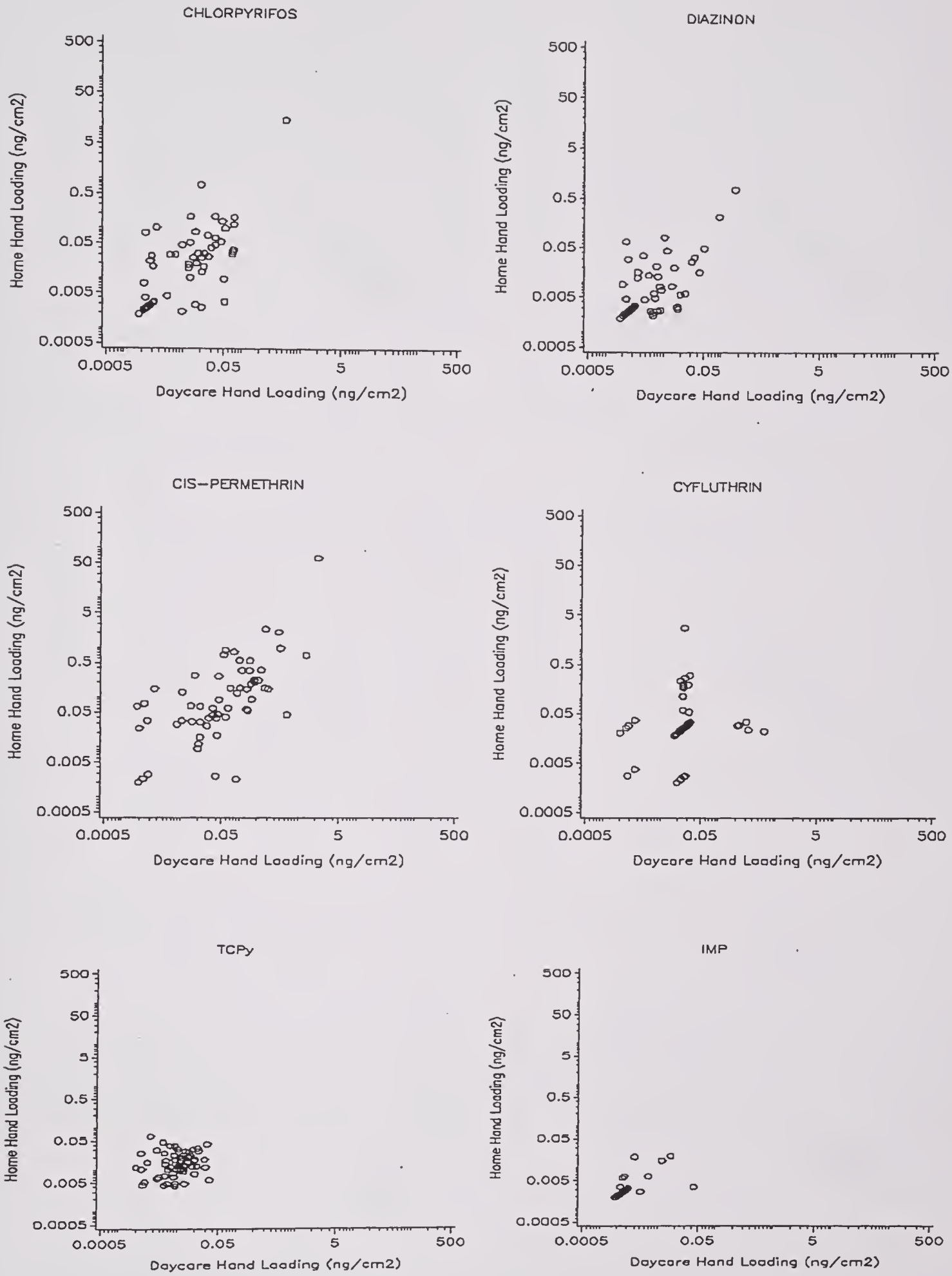


Figure 7.6 Relationship between children's hand loadings measured at CTEPP homes and daycares. Coefficients of determination (R^2) and slopes (β) for log (base 10) values: chlorpyrifos (R^2=0.47; β=0.91), diazinon (R^2=0.44; β=0.81), permethrin (R^2=0.41; β=0.72), cyfluthrin (R^2=0.02; β=0.19), TCPy (R^2=0.03; β=0.54), and IMP (R^2=0.31; β=0.54).

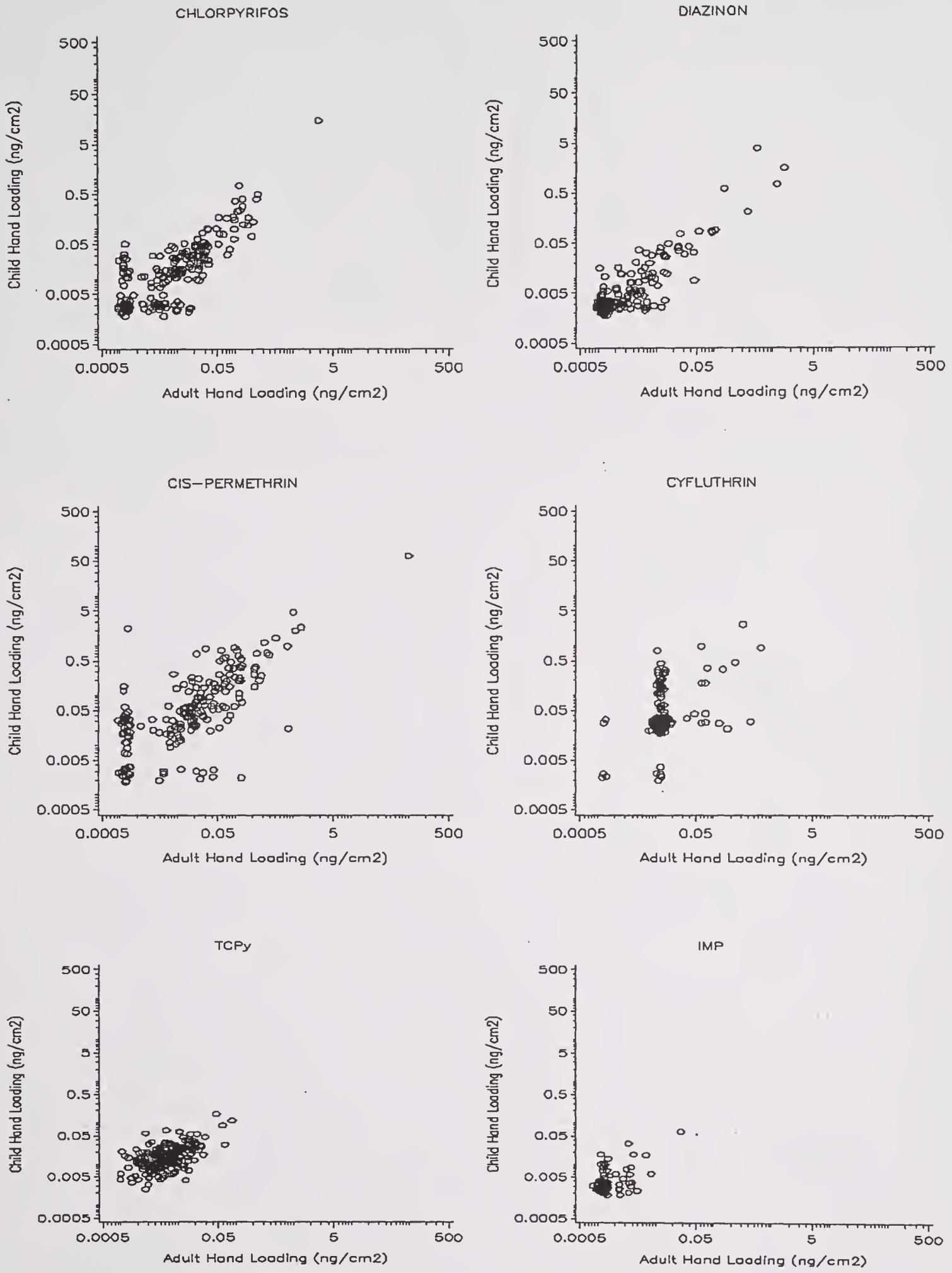
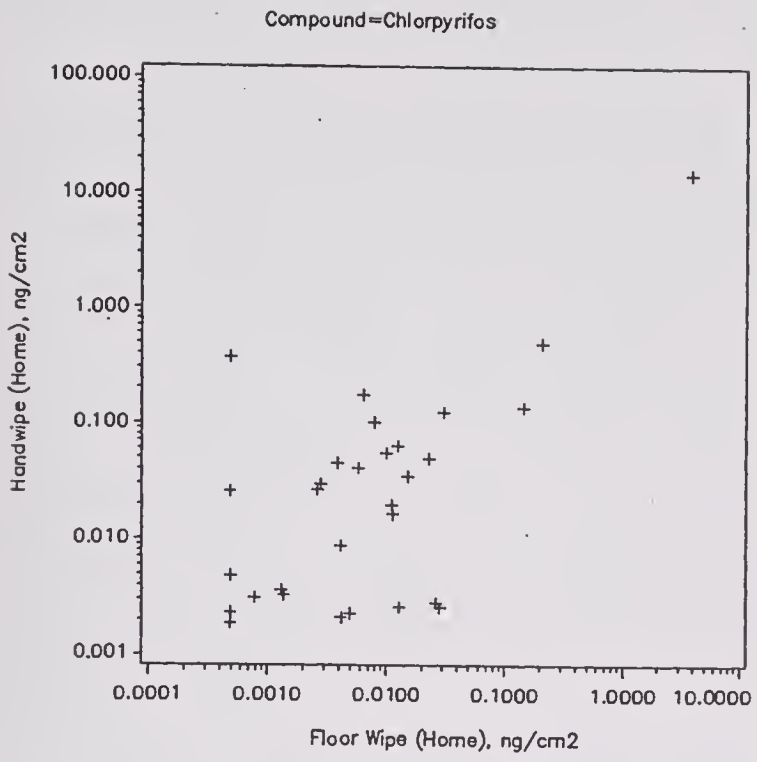
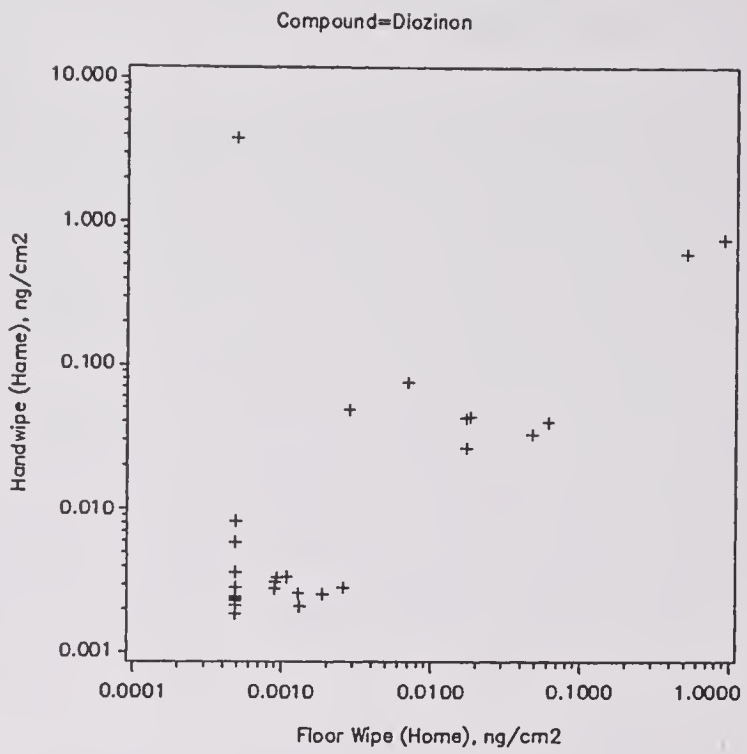
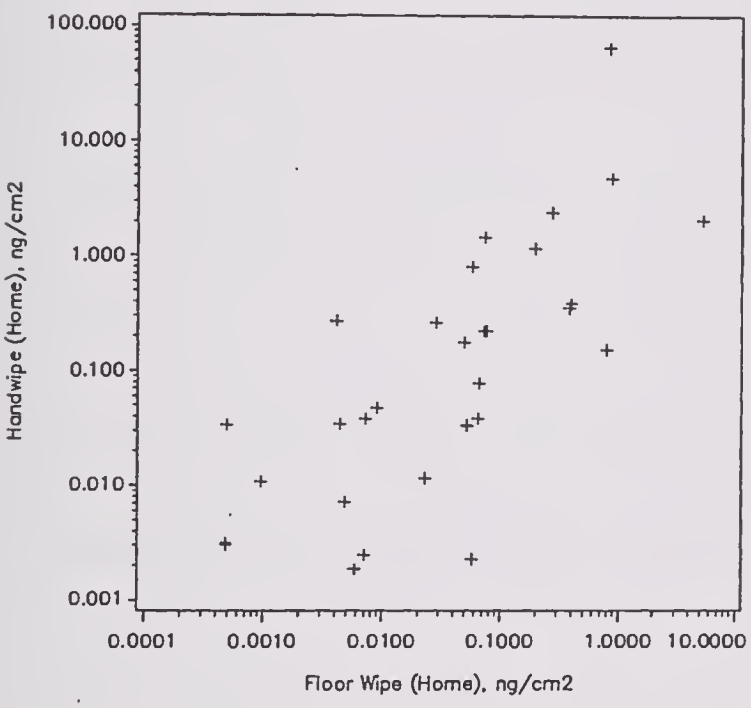


Figure 7.7 Relationship between hand loadings among children and adults in CTEPP. Coefficients of determination (R^2) and slopes (β) for log (base 10) values: chlorpyrifos (R^2=0.64; β=0.77), diazinon (R^2=0.77; β=0.81), permethrin (R^2=0.49; β=0.65), cyfluthrin (R^2=0.20; β=0.61), TCPy (R^2=0.30; β=0.47), and IMP (R^2=0.28; β=0.63).







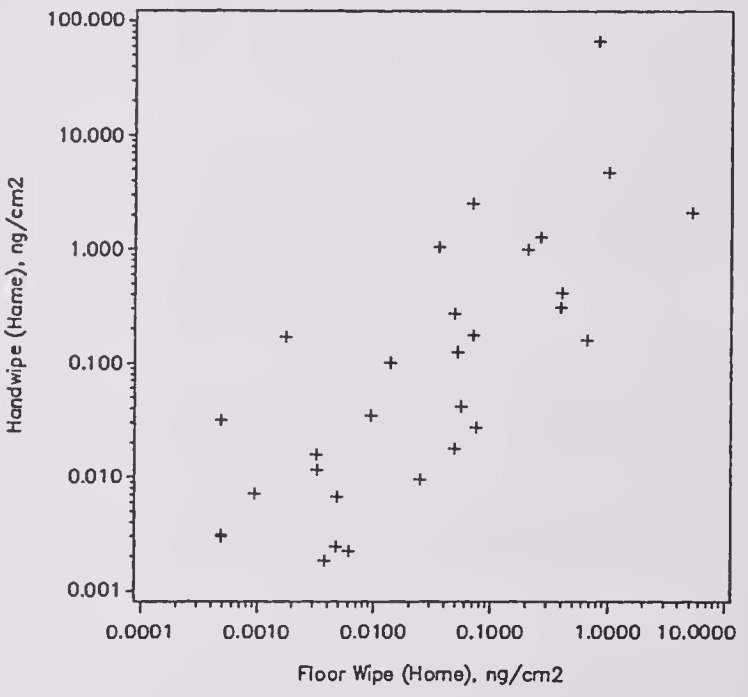


Figure 7.8 Relationship between hand wipe measurements and floor wipe measurements in CTEPP. Coefficients of determination (R^2) and slopes (β) for log (base 10) handwipe loadings regressed on log (base 10) floor wipe loadings are as follows: chlorpyrifos (R^2=0.38; β=0.64), diazinon (R^2=0.46; β=0.64), *cis*-permethrin (R^2=0.54; β=0.78), and *trans*-permethrin (R^2=0.60; β=0.82).

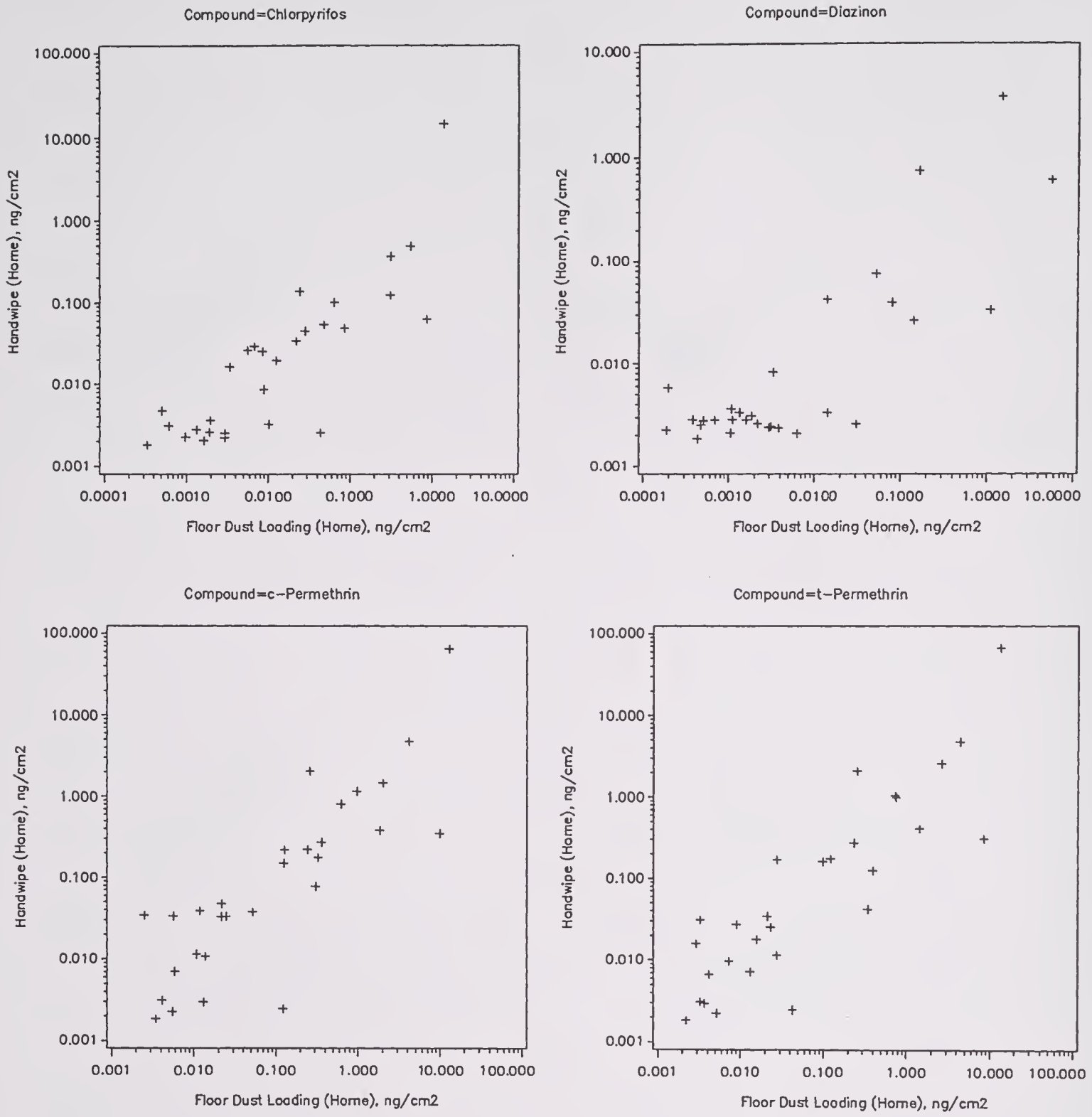


Figure 7.9 Relationship between hand wipe measurements and floor dust measurements in CTEPP. Coefficients of determination (R^2) and slopes (β) for log (base 10) handwipe loadings regressed on log (base 10) floor dust loadings are as follows: chlorpyrifos (R^2=0.71; β=0.78), diazinon (R^2=0.69; β=0.61), *cis*-permethrin (R^2=0.72; β=0.86), and *trans*-permethrin (R^2=0.76; β=0.88).

7.3 Measurements with Cotton Garments

The US EPA Office of Pesticide Programs uses a transfer coefficient approach to assess children's residential exposures to pesticides. The transfer coefficient approach was developed to assess occupational exposure in an agricultural setting, using empirically-derived dermal transfer coefficients to aggregate the mass transfer associated with a series of contacts with a contaminated medium. Dermal exposure sampling using a surrogate-skin technique such as a patch sampler or a whole-body garment sampler is conducted simultaneously with surface sampling for a specific activity, and a dermal transfer coefficient is then calculated. This transfer coefficient can then be used to estimate exposure for a similar activity by collecting only surface samples (Fenske, 1993), assuming that transfer is unidirectional (from surface to skin) and linear with time. Only limited research has been conducted to develop transfer coefficients for children in residential and daycare settings. Data were collected in the Daycare study (Cohen Hubal *et al.*, 2006), JAX, and CPPAES with cotton garments. The data are presented in Tables 7.10 to 7.12 and Figures 7.10 to 7.12.

- Comparison of mean chlorpyrifos loadings on socks in JAX and CPPAES (Table 7.10) with surface loadings (Table 4.4) suggests that higher surface loadings do not necessarily correspond to higher sock loadings across studies. It also suggests that perhaps activity levels influence transfer.
- The median chlorpyrifos loading on socks after a three-hour period in CPPAES was only about twice as high as the median loading after a one-hour period in the same environment (Table 7.10). This suggests that transfer to socks may not be linear with time, and again points towards the importance of activity levels.
- Bodysuit esfenvalerate loadings in the Daycare study were typically higher in the mornings, corresponding to higher group activity levels at that time (Figure 7.10). Depletion of surface loadings by morning activities is unlikely but was not tested.
- Multiple regression analysis of Daycare data suggests that body section (arms, legs, lower torso, and upper torso), relative activity level, and age group are all important predictors of bodysuit loadings (Table 7.11).
- The statistical significance of activity (Table 7.11), even when controlling for age group, suggests that activity level within age groups may be as important as age-related differences.
- The between- and within-person variability (GSD) in dermal exposures in the daycare setting (Table 7.12) is similar to what has been reported in agricultural/industrial settings.
- High within-person variability (compared to between-person variability) in cotton garment loadings (Table 7.12) suggests that factors related to changing environmental conditions and to differences in structured activities may be more important than child-specific characteristics.
- The relative standard deviations (%) of esfenvalerate loadings on cotton garment sections (Figure 7.11) were typically higher among infants during the morning sessions and among preschoolers during the afternoon sessions. This suggests that the structured

activities may have had a stronger influence on the observed variability than surface loadings in the respective rooms.

- Infants had 1.5 times as many hand wipe values (36%) above the MDL as preschool children (24%), consistent with the higher bodysuit loadings, perhaps reflecting greater contact with the floor surface. Figure 7.12 illustrates that among the hand wipes above the MDL, infants typically had higher loadings, with greater variability.
- The association between hand wipe samples above the limit of detection and average body suit loadings was statistically significant (Spearman rho = 0.54, $p < 0.05$, data not presented).

Table 7.10 Pesticide loading (ng/cm²) on cotton garments worn by children in three studies.

Study	Compound	Garment Type/Section	Age	N	% Det	MDL	Mean	SD	P50	P95
Daycare	Esfenvalerate	Arms	9-13 mo	26	92	0.01	0.12	0.18	0.06	0.42
			24-38 mo	28	100	0.01	0.1	0.09	0.07	0.23
		Legs	9-13 mo	26	100	0.01	0.27	0.21	0.22	0.75
			24-38 mo	28	93	0.01	0.2	0.41	0.1	0.46
		Lower Torso	9-13 mo	26	100	0.01	0.28	0.23	0.18	0.73
			24-38 mo	28	100	0.01	0.2	0.18	0.12	0.52
		Upper Torso	9-13 mo	26	96	0.01	0.05	0.05	0.03	0.12
			24-38 mo	28	100	0.01	0.09	0.13	0.05	0.16
CPPAES	Chlorpyrifos	Bottom	2-5 yr	7	100	0.01	0.58	0.37	0.7	1.0
		Knee	2-5 yr	14	100	0.01	0.62	0.4	0.7	1.2
		Leg	2-5 yr	14	100	0.01	0.38	0.27	0.45	0.8
		Sock (1 hr)	2-5 yr	14	100	0.01	8.6	14	3.5	53
		Sock (3 hr)	2-5 yr	14	100	0.01	10.8	13	7.6	30
JAX	Chlorpyrifos	Sock	4-6 yr	9	100	0.4	2.3	1.3	2.2	5.1
	Diazinon	Sock	4-6 yr	9	33	0.08	NC	NC	<0.08	1.8
	Esfenvalerate	Sock	4-6 yr	9	22	0.28	NC	NC	<0.28	2.6
	Cyfluthrin	Sock	4-6 yr	9	0	0.24	NC	NC	<0.24	<0.24
	cis-Permethrin	Sock	4-6 yr	9	44	0.8	NC	NC	<0.8	128
	trans-Permethrin	Sock	4-6 yr	9	100	0.2	23.6	59	1.44	180
CHAMACOS	Chlorpyrifos	Union Suit	6-10 mo	10	100	0.001	0.026	0.025	0.019	0.095
			21-27 mo	10	100	0.001	0.016	0.008	0.015	0.025
		Sock	6-10 mo	9	89	0.05	0.18	0.10	0.17	0.37
			21-27 mo	10	90	0.05	0.28	0.18	0.24	0.64
	Diazinon	Union Suit	6-10 mo	10	100	0.001	0.017	0.012	0.014	0.043
			21-27 mo	10	100	0.001	0.052	0.13	0.009	0.42
		Sock	6-10 mo	9	78	0.02	0.099	0.094	0.070	0.29
			21-27 mo	10	90	0.02	0.50	1.1	0.13	3.5
	Esfenvalerate	Union Suit	6-10 mo	10	10	0.02	NC	NC	<0.02	0.038
			21-27 mo	10	10	0.01	NC	NC	<0.01	0.047
		Sock	6-10 mo	9	11	0.25	NC	NC	<0.25	1.9
			21-27 mo	10	10	0.25	NC	NC	<0.25	2.3
	Cyfluthrin	Union Suit	6-10 mo	10	10	0.07	NC	NC	<0.07	1.1
			21-27 mo	10	0	0.04	NC	NC	<0.04	<0.04
		Sock	6-10 mo	9	0	2.5	NC	NC	<2.5	<2.5
			21-27 mo	10	10	2.5	NC	NC	<2.5	14
	cis-Permethrin	Union Suit	6-10 mo	10	100	0.001	0.19	0.11	0.18	0.41
			21-27 mo	10	100	0.001	0.96	2.4	0.16	7.9
		Sock	6-10 mo	9	100	0.02	2.0	2.8	1.1	8.7
			21-27 mo	10	100	0.02	6.2	13	1.8	43
	trans-Permethrin	Union Suit	6-10 mo	10	100	0.001	0.18	0.35	0.088	1.2
			21-27 mo	10	100	0.001	0.96	2.6	0.059	8.4
		Sock	6-10 mo	9	100	0.02	2.6	2.4	1.9	7.7
			21-27 mo	10	100	0.02	10	22	2.0	71

NC, Not calculated

Table 7.11 Results of multiple linear regression modeling of measured bodysuit pesticide loading ($ng/cm^2/sec$) from data collected in the daycare study.

Effect	Level	Estimate	p-Value
Intercept	intercept	-1.43	<0.0001
Bodysuit Section	arms	0.46	<0.0001
	legs	1.05	
	lower torso	1.35	
	upper torso	0	
Visit	first	0.87	0.0006
	second	0.31	
	third	0	
Session	am	0.44	0.0006
	pm	0	
Activity Level	high	1.36	<0.0001
	middle	0.65	
	low	0	
Classroom	infant	0.38	0.0386
	preschool	0	

Table 7.12 Estimates of between- and within-person variability for loading on individual bodysuit sections.

Statistic	Arms	Upper	Legs	Lower
Between-person variance (logged)	0.26	0.04	0.67	0.37
Within-person variance (logged)	0.76	0.76	1.02	0.59
Intraclass Correlation Coefficient	0.25	0.05	0.40	0.39
GSD, between	1.7	1.2	2.3	1.8
GSD, within	2.4	2.4	2.7	2.2

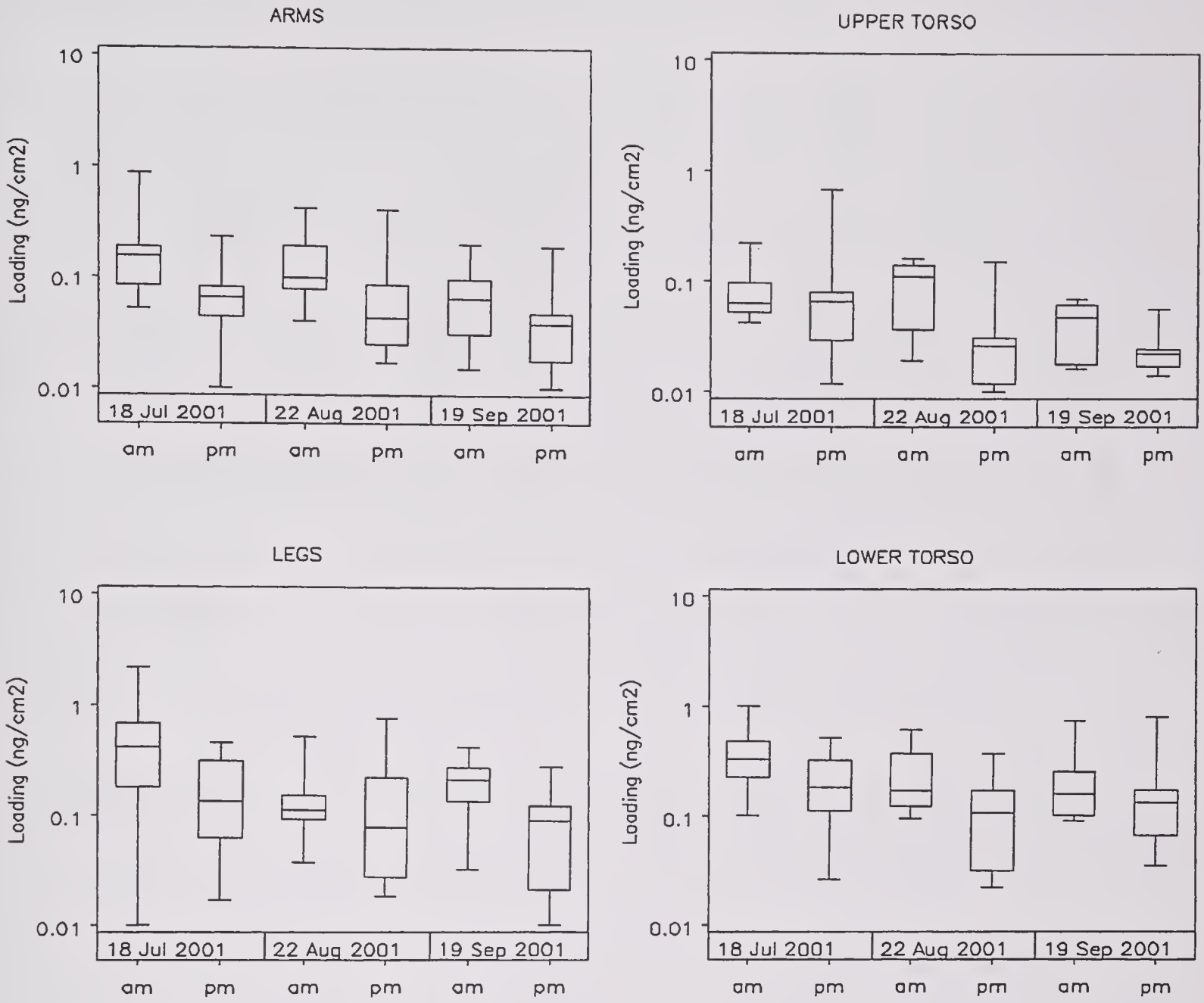


Figure 7.10 Bodysuit section loadings (ng/cm²) by monitoring period from the Daycare study.

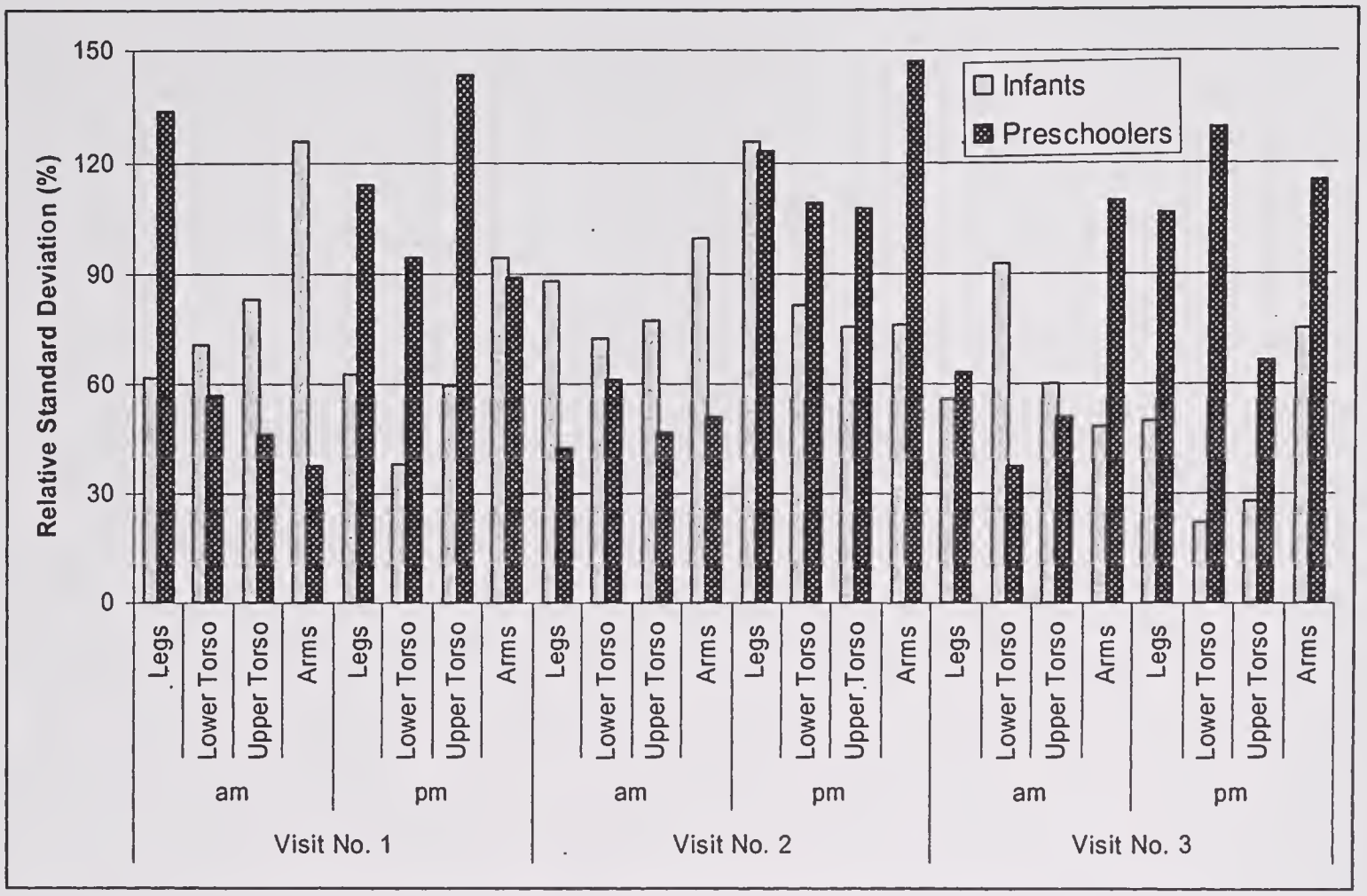


Figure 7.11 Relative standard deviations of esfenvalerate loadings on cotton garment sections among infants and preschoolers in the Daycare study.

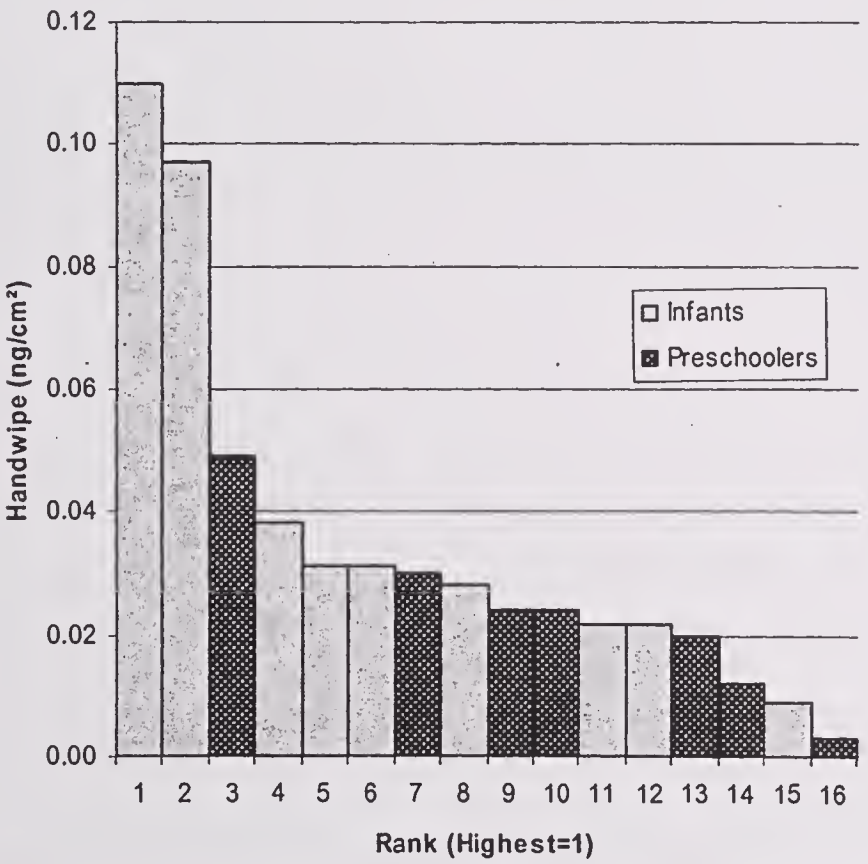


Figure 7.12 Handwipe loadings (ng/cm²) above method detection limit among infants and preschoolers in the Daycare study. Values are sorted in descending order, illustrating that the highest loadings were typically from infants and the lowest typically from preschoolers.

8.0 URINARY BIOMARKER MEASUREMENTS

Biological markers are indicators of the actual body burden of a chemical. As such, they reflect all routes of exposure, as well as inter-individual differences in absorption and metabolism. Moreover, they are often more directly related to potential adverse health effects than the external concentrations (Lowry, 1986: Hulka and Margolin, 1992). In human observational measurement studies involving young children, urine is the primary vehicle for biomonitoring. Urine is advantageous over blood because of its noninvasiveness, ease of collection, and large available quantity. Urinary biomarkers, however, also have disadvantages related to uncertainties in the fraction of the absorbed compound that is eliminated and in the precision of the measurements.

The relationship between a biological marker and external exposure is influenced by factors related both to the environment and to human physiology. Factors related to the environment include spatial and temporal variability in exposure concentrations (as discussed in earlier chapters of this report) and effects of the presence of other chemicals (Coble et al, 2005). Factors related to human physiology include differences, both over time and across individuals, in the rates of absorption, distribution, metabolism, and excretion (Droz, 1989). When biological monitoring and exposure monitoring are used together, the relationship between the two may be evaluated to investigate the relative contribution of the various exposure routes.

Evaluating the relative contribution of exposure routes to aggregate intake is subject to error related to estimates of exposure and of aggregate intake. Issues related to route-specific exposure estimates have been discussed earlier. Dependable information on the toxicokinetics (absorption, distribution, metabolism, and excretion) of a compound are necessary for reliable estimates of aggregate intake, whether those estimates are derived from the sum of route-specific absorption estimates or from excreted biomarker levels. To accurately estimate aggregate intake from excreted biomarker levels, urinary biomarker output rates must be calculated from the biomarker levels. Such calculations require information on the entire urine volume and elapsed time since previous void - information that has rarely been collected in field studies.

8.1 Toxicokinetics of Organophosphate and Pyrethroid Pesticides

Some understanding of organophosphate and pyrethroid pesticide toxicokinetics is necessary to meaningfully compare the environmental and dietary concentrations presented in the previous chapters with the urinary biomarker concentrations presented in this chapter. Despite extensive usage, remarkably little is available from the scientific literature on kinetic parameters in humans. Parameters reported for absorption of parent compounds and elimination of urinary metabolites following pesticide exposure are summarized in Table 8.1.

Absorption

Inhalation studies with a variety of gases have shown that even the most efficiently absorbed low molecular weight, highly water soluble compounds rarely exceed 70% uptake. No studies reporting the fraction of organophosphate pesticides absorbed through inhalation were found, but

Leng *et al.* (1997) reported that only about 16% of the pyrethroid cyfluthrin was absorbed through inhalation.

The importance of the dietary contribution to aggregate exposure among infants and young children is well known (NRC, 1993), but few studies have investigated what fraction of ingested pesticide residue is absorbed. For organophosphates, Nolan *et al.* (1984) estimated 70% absorption of chlorpyrifos based on urinary 3,5,6-trichloro-2-pyridinol (TCPy), whereas others estimated 60% to 93% absorption based on dialkylphosphate (DAP) metabolites (Garfitt *et al.*, 2002; Griffin *et al.*, 1999). Diet reportedly affects absorption (Timchalk *et al.*, 2002). As for pyrethroids, Woollen *et al.* (1992) estimated that 27-57% of cypermethrin was absorbed, while Eadsforth and colleagues (1983; 1988) estimated 45-49% and 72-78% for the *cis* and *trans* isomers, respectively.

Dermal absorption is typically low due to loss by washing, evaporation, or exfoliation (Feldmann and Maibach, 1974). For organophosphate pesticides, absorption of chlorpyrifos was estimated, based on its primary metabolite TCPy, to be 1.28% of an applied dose of 4 mg/cm² (over 12-20 hr) (Nolan *et al.;* 1984), and 1.2% and 4.3% of applied doses of 0.15 and 0.05 mg/cm² (over 4 hr), respectively (Meuling *et al.*, 2005). Absorption of both chlorpyrifos and diazinon was estimated to be about 1% of applied doses of about 0.4 and 1.3 mg/cm² (over 8 hr), respectively, based on DAP metabolites (Griffin *et al.,* 1999; Garfitt *et al.*, 2002). The percent that is absorbed increases as the applied dose (per cm²) decreases. Large differences have been reported by anatomical area (Maibach *et al.*, 1971) and among individuals (Feldmann and Maibach, 1974). For pyrethroids, Bartelt and Hubbell (1987) found only about 2% of applied permethrin to be absorbed within 24 h. Wester *et al.* (1994) observed that approximately 2% (forearm) and 7.5% (scalp) of radiolabeled pyrethrin was absorbed. The ATSDR (2001) has concluded that for pyrethroids in general, < 2% of the applied dermal dose is absorbed, at a rate much slower than that by the oral or inhaled routes.

Due to the paucity of available information on absorption from human studies, simple default values based on human studies, animal studies, and conservative assumptions are often required. For small children (ages 1-6) the following route-specific absorption is often assumed: 50-100% for inhalation, 50% for ingestion, and 1-3% for dermal. In addition, a daily intake of 100 mg of house dust is assumed for indirect ingestion. These absorption assumptions are a source of substantial uncertainty in route-specific intake estimates. In fact, since dermal absorption increases with decreasing dermal loadings (as demonstrated above with organophosphates), default assumptions of less than 3% for dermal absorption may underestimate absorption at the very low levels measured in field studies

Distribution and Metabolism

Once in the bloodstream, organophosphate or pyrethroid pesticides are rapidly distributed and metabolized. A typical organophosphate (OP) pesticide is composed of a dialkyl (either dimethyl or diethyl) phosphate moiety and an organic group. Hydrolytic cleavage of the ester bond yields one dialkylphosphate (DAP) metabolite and one organic group moiety (Barr *et al.,* 2004). Dimethyl OPs (including malathion, phosmet, and azinphos-methyl) produce dimethyl metabolites and diethyl OPs (including chlorpyrifos and diazinon) produce diethyl metabolites

(Aprea *et al.,* 2002). The organic group metabolites, including 2-isopropyl-6-methyl-4-pyrimidinol (IMPy) for diazinon and 3,5,6-trichloro-2-pyridinol (TCPy) for chlorpyrifos, are considered to be semi-specific.

Pyrethroids are esters of chrysanthemic acid and benzyl alcohols. Hydrolytic cleavage of the ester bond yields a benzoic acid and a chrysanthemic acid derivative. The 3-phenoxybenzoic acid (3-PBA) metabolite is common to 10 of the 18 pyrethroids registered in the United States including permethrin, cypermethrin, deltamethrin, esfenvalerate (Baker *et al.,* 2004). Other benzoic acid metabolites analogous to 3-PBA are more specific and include 4-fluoro-3-phenoxybenzoic acid (4F3PBA) from cyfluthrin and 2-methyl-3-phenylbenzoic acid (MPA) from bifenthrin. These are not necessarily terminal metabolites; for example, as much as 38% of 3-PBA has been reported by Woollen *et al.* (1992) to undergo further oxidation to 3-(4'-hydroxyphenoxy) benzoic acid (4OH3PBA). The chrysanthemic acid derivative *cis*-2,2-dibromovinyl-2,2-dimethyl-cyclopropane-1-carboxylic acid (DBCA) is specific to deltamethrin while the *cis*- and *trans*- isomers of 2,2-dichlorovinyl-2,2-dimethyl- cyclopropane-1-carboxylic acid (DCCA) are common to permethrin, cypermethrin, and cyfluthrin.

Excretion

Both the OPs and the pyrethroids are rapidly eliminated in urine. Elimination appears to follow first-order kinetics, with elimination half-times in humans ranging from 2 to 41 hours for OPs and from 6.4 to 16.5 hours for pyrethroids, depending on both the compound and the route of exposure (ATSDR, 2001; Garfitt *et al.*, 2002; Meuling *et al.*, 2005). The elimination half-life of about 8 hours reported for 3-PBA among workers exposed to cypermethrin (Kuhn *et al.*, 1999) suggests that 88% of the metabolite is excreted within the first 24 hours following exposure.

Route-specific differences in the peak excretion of urinary OP pesticide metabolites have been reported (Griffin *et al.*, 1999; Garfitt *et al.*, 2002; Meuling *et al.*, 2005). Peak excretion is observed to occur 6 to 24 hours later when absorption is by the dermal route compared to when absorption is by the oral route, largely because of route-specific differences in absorption. Peak excretion may occur as late as 48 hours following dermal exposure, as observed among volunteers performing scripted "Jazzercise" activities (Krieger *et al.*, 2000). Extended peak excretion times suggest that chlorpyrifos may be retained by the skin and may remain systemically available for prolonged periods (Meuling *et al.*, 2005)

While the above toxicokinetic studies evaluate excreted mass or mass rates, our past field studies have largely evaluated only biomarker concentrations. In the future, all studies should include information on void volumes and times to allow excreted mass to be calculated. Relevant transformations can be found in Rigas *et al.* (2001) and are currently incorporated in the SHEDS model.

Table 8.1 Absorption and elimination characteristics for pesticides and urinary biomarkers of pesticide exposure.

COMPOUND	ABSORPTION OF PARENT COMPOUND			ELIMINATION OF METABOLITES		
	ORAL	DERMAL	INHALATION	ORAL	DERMAL	INHALATION
Chlorpyrifos	Volunteer studies: 70% of oral dose excreted in urine as TCPy (Nolan *et al.*, 1984), 93% of oral dose excreted in urine as dialkyl-phosphates (Griffin *et al.*, 1999). Absorption factor estimated at 0.90 (ATSDR).	Volunteer studies: 1.3% of dermal dose excreted in urine as TCPy (Nolan *et al.*, 1984). 1% of dermal dose excreted as dialkyl-phosphates (Griffin *et al.*, 1999), 1.2 – 4.3% of dermal dose excreted as TCPy (Meuling *et al.*, 2005).	No Information.	Volunteer study, 27 h oral (Nolan *et al.*, 1984). Volunteer study, approx 15.5 h oral (Griffin *et al.*, 1999).	Volunteer study, 27 h dermal (Nolan *et al.*, 1984). Volunteer study, approx 30 h dermal (Griffin *et al.*, 1999). Volunteer study, approx 41 h dermal (Meuling *et al.*, 2005).	No Information.
Diazinon	Human oral absorption approx. 60% (Garfitt *et al.*, 2002). Default oral absorption factor of 0.85 (ATSDR).	Human dermal absorption rate: 456 ng/cm²/h (Garfitt *et al.*, 2002).	No Information.	Human study, 2 h oral (Garfitt *et al.*, 2002).	Human study, 9 h dermal (Garfitt *et al.*, 2002).	No Information.
Pyrethroids (as a group)	Absorption is incomplete, minimum estimate 40 - 60%, but first- pass metabolism may underestimate absorption (ATSDR, 2001).	<2% of the applied dermal dose is absorbed, rate of absorption much slower than by the oral or inhaled routes; may be stored in skin and then slowly released into the systemic circulation (ATSDR, 2001).	Rapidly absorbed in humans following inhalation, but no estimates of fraction absorbed are available (ATSDR, 2001).	Elimination appears to follow first-order kinetics, with elimination half-times in humans ranging from 6.4 to 16.5 hours, depending upon the specific pyrethroid and exposure route studied (ATSDR, 2001).		
Permethrin	Oral absorption factor of 0.70 suggested (NRC).	Poor dermal absorption: ~2% of applied dose absorbed/24 h (Bartelt and Hubbell, 1987); 7.5% (scalp) and 1.9% (forearm) of applied dose (Wester *et al.*, 1994).	No Information.	No Information.	No Information.	No Information.
Cyfluthrin	No Information.	No Information.	Human data suggest ~15% absorption (Leng et al, 1997).	Human oral dosing produced t-½ of 6.4 h (Leng et al, 1997b).	No Information.	Human ½-lives of 6.9 h (c-DCCA), 6.2 h (t-DCCA), 5.3 h (FPBA) (Leng et al, 1997).
Cypermethrin	Human volunteer study 27-57% (mean 36%) cypermethrin absorbed (Woollen *et al.*, 1992).	No Information.	No Information.	Human oral dosing, urinary metabolites have mean ½-life of 16.5 h (Woollen *et al.*, 1992).	Human dermal dosing, excretion rates peaked at 12-36 h, mean ½-life was 13 h (Woollen *et al.*, 1992).	No Information.

8.2 Measurements of Pesticide Metabolites in Urine

Urinary biomarkers were measured in several large-scale and pilot-scale children's observational measurement studies described in Table 8.2. These include the MNCPES, CTEPP, NHEXAS-AZ, CPPAES, JAX, CHAMACOS, PET, and DIYC studies. All urine samples were collected exclusively at the children's homes except for the CTEPP study, in which urine samples were also collected at daycare centers. Urine collection followed outdoor turf applications in the PET study and routine professional indoor applications in the DIYC and CPPAES studies.

Spot urine samples, mainly first morning voids, were collected using age-appropriate methods including under-toilet seat bonnets (CTEPP, PET), collection cup (NHEXAS-AZ, MNCPES), diaper insert (DIYC), and "potty chair" (CPPAES). Table 8.3 presents selected organophosphate (OP) and pyrethroid metabolites that were measured in the children's urine samples in multiple studies. The pesticide metabolites are 3,5,6-trichloro-2-pyridinol (TCPy), 2-isopropyl-6-methyl-4-pyrimidinol (IMP), and 3-phenoxybenzoic acid (3-PBA).

Sample collection was performed by the children's caregivers following protocols provided by the investigators. Chemical analysis of urinary metabolites in nearly all included studies was performed by the National Center for Environmental Health of the Centers for Disease Control and Prevention in Atlanta, GA, using validated tandem mass spectroscopy techniques (Baker *et al.,* 2000; Baker *et al.,* 2004; Beeson *et al.,* 1999; Hill *et al.,* 1995). Chemical analysis for the CTEPP study was performed by Battelle Institute using validated gas chromatography/mass spectroscopy techniques.

Limits of detection for each pesticide metabolite are given by study in Table 8.4. Detection frequencies are provided in Figure 8.1. Concentrations for the median and 95th percentiles for each urinary metabolite are presented by study in Table 8.5. Figure 8.2 shows the log probability plots of urinary TCPy and 3-PBA concentrations for children across large observational field studies. Figure 8.3 presents the box-and-whisker plots that graphically depict the urinary TCPy and 3-PBA concentrations for both the large-scale and pilot-scale children's observational measurement studies.

The National Health and Nutrition Examination Survey (NHANES) includes an ongoing assessment of the exposure of the U.S. population to environmental chemicals through the measurement of biomarkers. Spot measurements of urinary pesticide biomarkers among children 6 to 12 years old from both the 1999-2000 and the 2001-2002 cycles are included for comparison with results from our studies. Please note that NHANES does not report results by region or by season.

- The chlorpyrifos metabolite TCPy was detected in over 90% of the children's urine samples in all listed studies. The pyrethroid metabolite 3-PBA was detected in over 60% of the CTEPP-OH samples and over 90% of the JAX samples (Figure 8.1).
- The urinary TCPy concentrations were at least an order of magnitude higher than the urinary 3-PBA concentrations across studies (Figure 8.2).

- There is virtually no difference in urinary TCPy concentrations measured in CTEPP NC and OH, but the concentrations from Minnesota and Arizona are substantially higher (Figure 8.2, all unweighted). Higher levels in MNCPES and NHEXAS-AZ may reflect intentional oversampling of pesticide-using households in MNCPES, and greater use of chlorpyrifos at the time that MNCPES and NHEXAS-AZ were conducted.
- Compared to values for children under 12 years old collected in the 1999-2002 NHANES (Figure 8.2), the median TCPy values were higher in all of our studies, but the 95th percentile values were only higher for MNCPES.
- The children in JAX had levels of 3-PBA that were at least seven times higher than those of children in CTEPP-OH (Figure 8.3). All urine data from JAX participants suggest that JAX is a high pesticide usage area.
- The median 3-PBA value in CTEPP (0.3 ng/mL) was similar to NHANES (0.3 ng/mL), but the median JAX value (2.2 ng/mL) was much higher (Figure 8.3).
- Levels of IMP were about an order of magnitude higher in DIYC compared to PET or NHANES (Figure 8.3).
- The median urinary TCPy concentration was the highest for the NHEXAS-AZ and JAX studies and the lowest for the CTEPP-NC and CTEPP-OH studies (Table 8.5).
- In the CPPAES study, the intensity of the crack and crevice applications of chlorpyrifos was described as either high (n = 7) or low (n = 3), with mean air concentrations resulting from "high" applications five orders of magnitude higher than those from "low" applications. Figure 8.4 shows that the urinary TCPy concentrations over time were not much different for the children in the high versus low application groups.
- For children in the "high" application group in CPPAES, the median urinary TCPy concentration one day before application of chlorpyrifos was higher than on the first two days following application (Figure 8.4). Crack and crevice applications of chlorpyrifos at these homes did not substantially increase the children's urinary TCPy concentrations.
- The concentration-time profiles for urinary TCPy levels in CPPAES did not mirror the environmental concentration time profiles (Figure 8.5).

Table 8.2 Summary of the children's urinary biomarker collection methods.

Study	N	Age Range	Sampling Device	Collection Strategy	Collection After Pesticide Use	Collection Frequency	Analytes [a]	Urinary Output Correction Factors
NHEXAS-AZ (subset)	21	5 to12 yr	Urine collection cup	Morning void	No	Once (in 3-day monitoring period)	TCPy	Creatinine
MNCPES	102	3 to 13 yr	Urine collection cup	Morning void	No	Days 3, 5, and 7 of sampling period	TCPy	Creatinine
CTEPP	257	2 to 5 yr	Bonnet for children, urine collection cup for adults	Morning void, after lunch, after dinner/ before bedtime	Only for some homes (~15%)	Over a 48-hr period	TCPy, 3-PBA (Ohio, only)	Creatinine, Specific gravity
JAX	9	4 to 6 yr	Plastic cup	Morning void	Yes, indoor	1 day	TCPy, IMP, 3-PBA	Creatinine
CHAMACOS	20	6 to 24 mo	Cotton diaper and Infant urine collection bag or commode container	One overnight and one spot sample	No	Once	Dialkyl Phosphate metabolites	Creatinine
CPPAES	10	2 to 4 yr	Toys R' Us child's potty, plastic cup	Morning void	Yes, indoor	Pre- and days 1, 2, 3, 5, 7, 9, and 11 post-application	TCPy	Creatinine
PET Pilot	6	5 to 12 yr	Urine collection bottle or urine bonnet	Morning void	Yes, outdoor	Pre- and days 1, 2, 4, and 8 post-application	IMP	Creatinine
DIYC	3	1 to 3 yr	Diaper insert or collection cups	Morning void and other spot samples	Yes, indoor	Days 3, 5, and 7 post-application	IMP	Creatinine

[a] Analytes relevant to interstudy comparison. Most studies included additional metabolites.

Table 8.3 Urinary metabolites of organophosphate and pyrethroid pesticides measured in the children's observational measurement studies.

Metabolite	Parent Compound
3,5,6-Trichloro-2-pyridinol (TCPy)	Chlorpyrifos [a]
2-Isopropyl-6-methyl-4-pyrimidinol (IMP)	Diazinon
3-Phenoxybenzoic acid (3-PBA)	Permethrin [b]

[a] TCPy is also a metabolite of chlorpyrifos-methyl, which may occur in children's diet.
[b] Several other pyrethroids are metabolized into 3-PBA including cypermethrin, deltamethrin, fenvalerate, fluvalinate, permethrin, sumithrin.

Table 8.4 Limits of detection (ng/mL) for each pesticide metabolite measured in the children's urine samples by study.

Study	TCPy	IMP	3-PBA
NHEXAS-AZ	1.0	NA	NA
MNCPES	1.4	NA	NA
CTEPP-NC	1.0	NA	NA
CTEPP-OH	1.0	NA	0.2
JAX	0.4	2.0	0.5
CPPAES	1.0	NA	NA
PET	NA	0.3	NA
DIYC	NA	1.0	NA

NA, Not Applicable.

Table 8.5 Median and 95th percentile values (ng/mL) for the pesticide metabolites TCPy, IMP, and 3-PBA measured in the children's urine samples by study.

Study	TCPy		IMP		3-PBA	
	P50	P95	P50	P95	P50	P95
NHEXAS-AZ	12.0	26.0	NA	NA	NA	NA
MNCPES	7.2	23.0	NA	NA	NA	NA
CTEPP-NC	5.3	15.5	NA	NA	NA	NA
CTEPP-OH	5.1	12.3	NA	NA	0.3	1.8
JAX	9.8	21.2	<MDL	<MDL	2.2	98.7
CPPAES	7.7	18.0	NA	NA	NA	NA
PET	NA	NA	0.71	6.58	NA	NA
DIYC	NA	NA	7.1	27.0	NA	NA

NA, Not Applicable.

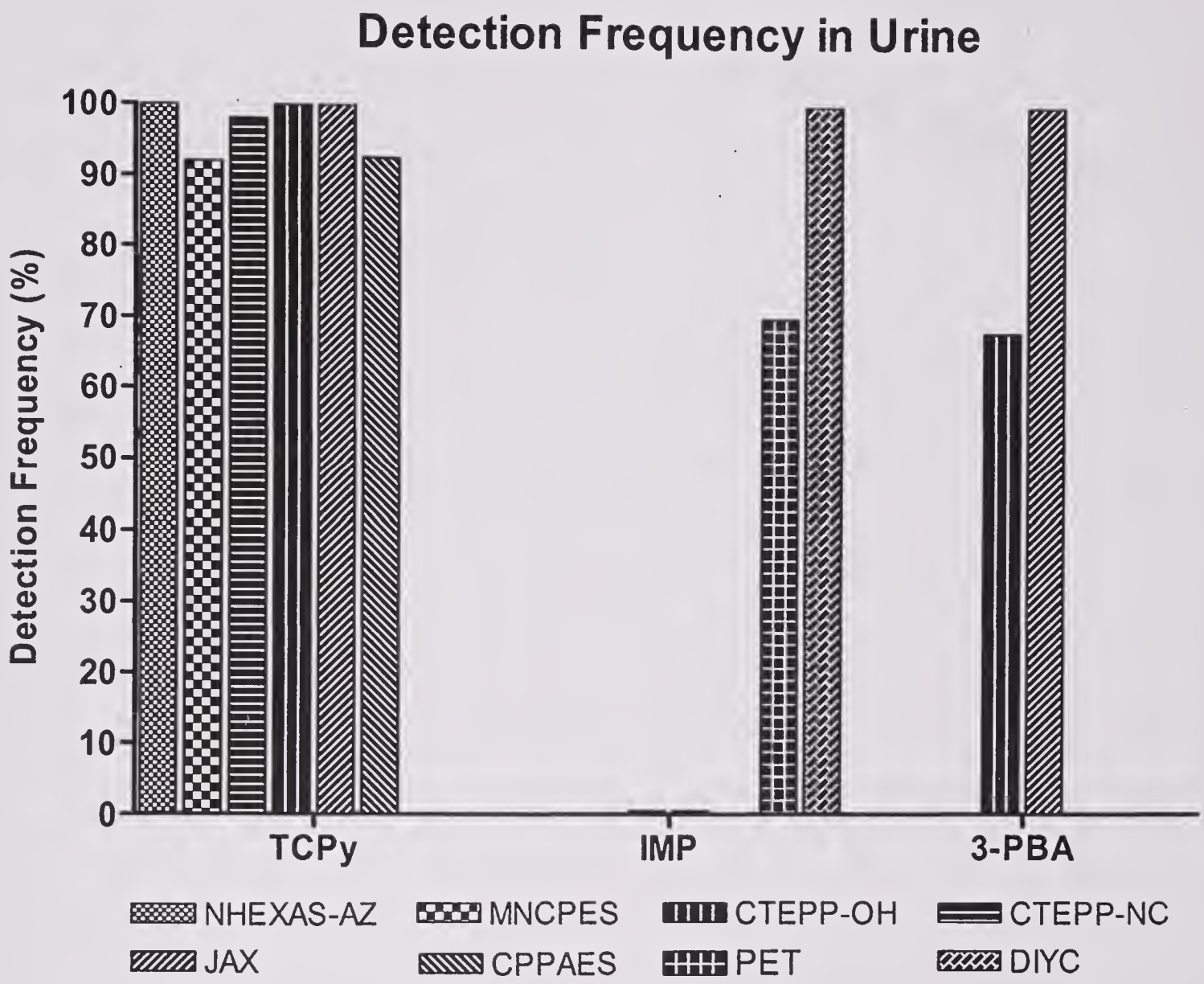


Figure 8.1 Detection frequencies of pesticide metabolites in the children's urines samples by study.

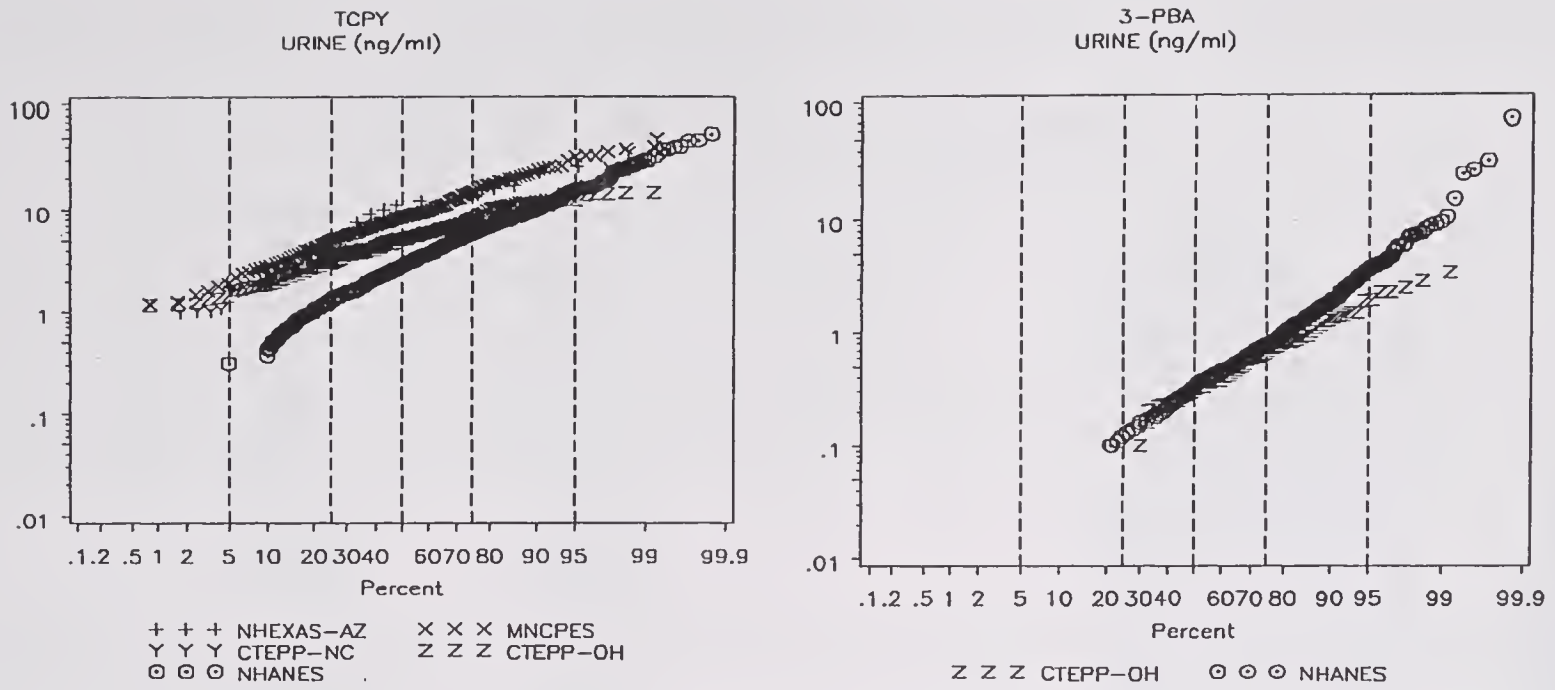
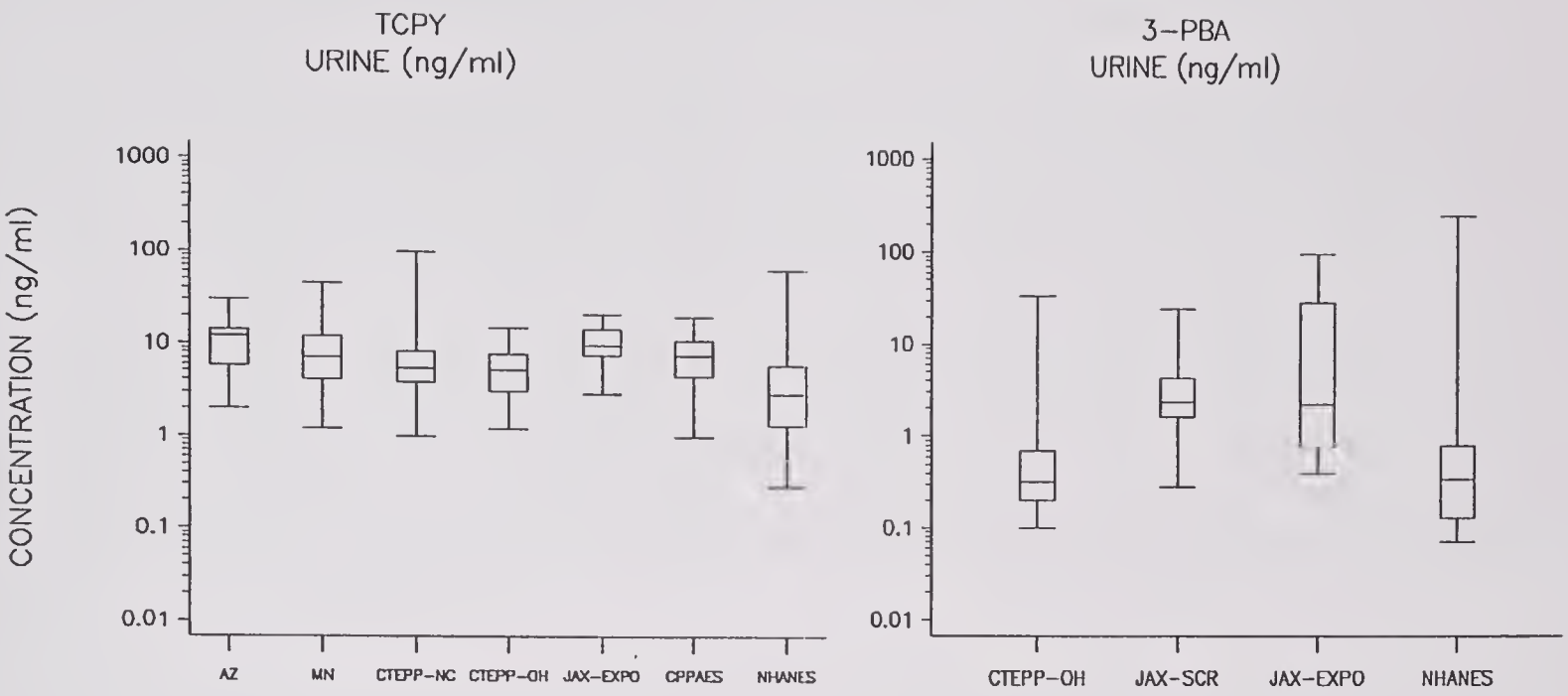


Figure 8.2 Log probability plots of urinary TCPy, 3-PBA, and IMP concentrations across large observational field studies. NHANES results are included for comparison.



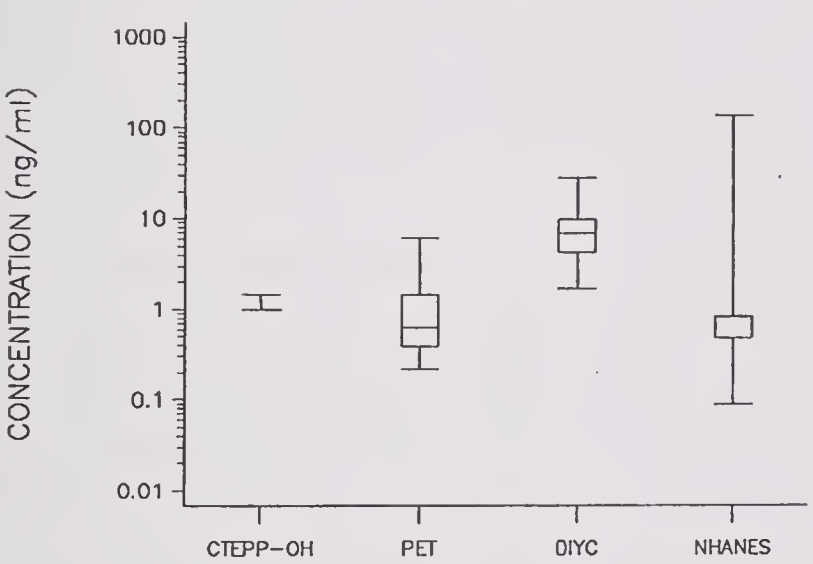


Figure 8.3 Box-and-whisker plots comparing the urinary TCPy and 3-PBA concentrations across studies.

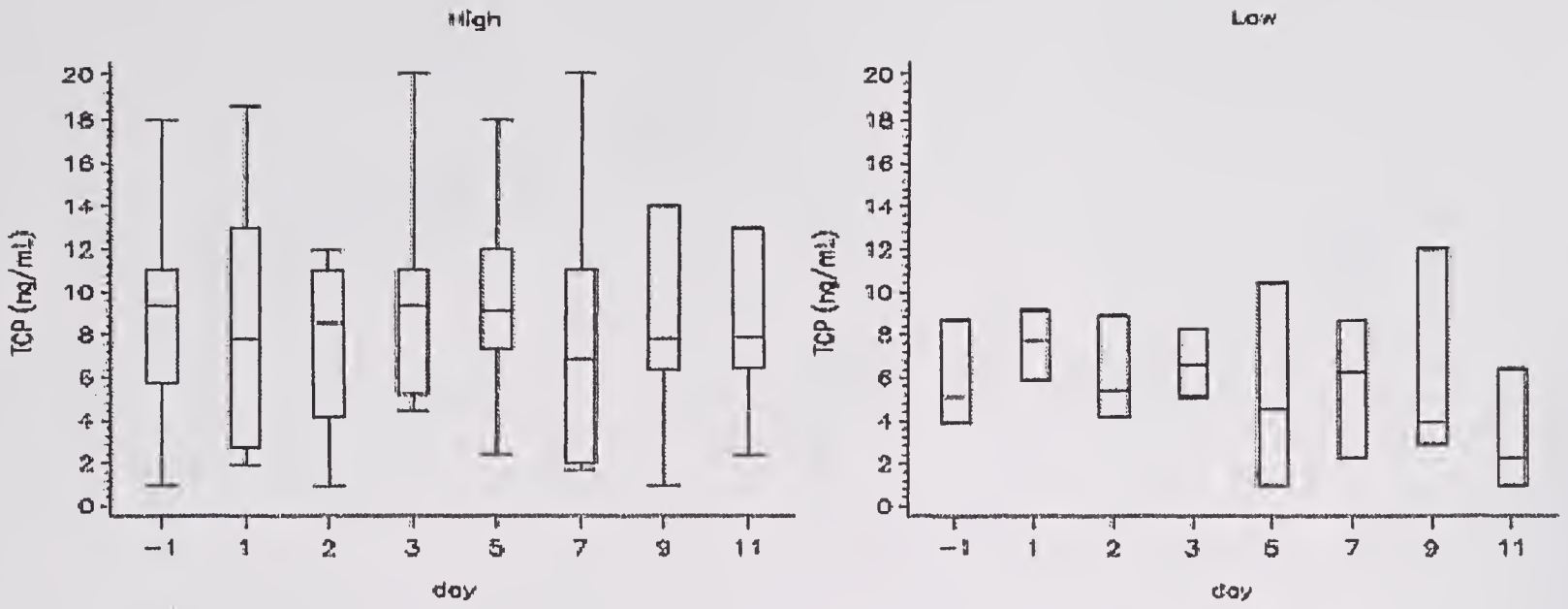


Figure 8.4 Urinary TCPy concentrations (ng/mL) over time for the children in the high and low application groups in CPPAES.

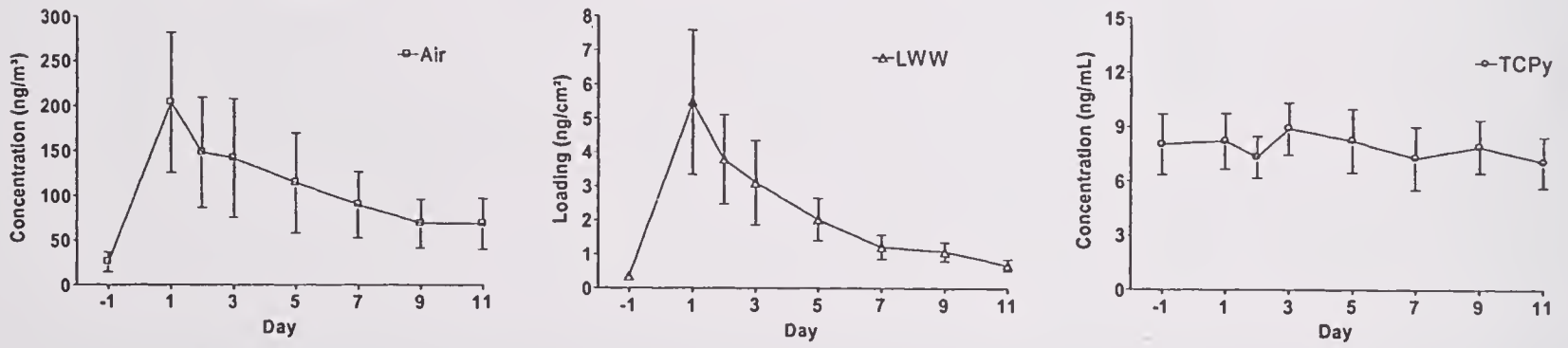


Figure 8.5 Time profiles for chlorpyrifos in environmental media and TCPy concentrations in urine for all children in the CPPAES.

8.3 Temporal Variability in Biomarker Measurements

In the CTEPP study, the children's spot urine samples (up to six per child) were analyzed separately for pesticide metabolites if the participants reported that a pesticide had been used in their homes within seven days of field monitoring. Figure 8.6 shows the variability of urinary TCPy concentrations in the children's urine samples over a 48-h period.

Intraclass correlation coefficients (ICCs) for urinary TCPy and 3-PBA concentrations in NC and OH children in the CTEPP study are provided in Table 8.6. The between and within-person geometric standard deviations (GSDs) for logged urinary concentrations of TCPy and 3-PBA for the NC and OH children in the CTEPP study are given in Table 8.7. Concentration-time profiles for TCPy and 3-PBA among CTEPP children are provided in Figure 8.6 and for IMP among PET study children in Figure 8.7.

- Relatively low ICCs (Table 8.6) indicate that a single measurement may not adequately represent the mean of the 48-hr sampling period for 3-PBA among adults and TCPy among children. Consistency of urinary metabolite concentrations over even short periods of time appears to be dependent on both the metabolite and the study population.
- Within-person GSDs are equal to or nearly equal to between-person GSDs for both TCPy and 3-PBA in urine measured in CTEPP (Table 8.7). This indicates that a single spot urine measurement is not sufficient to differentiate among children over a 48-hr time frame.
- Spot urine measurements over 48 hours among CTEPP participants reporting recent pesticide applications show large sample-to-sample variability and large differences among individuals (Figure 8.6).
- Adjustment of urinary metabolite values by specific gravity did not meaningfully reduce within-person variability of TCPy (Figure 8.6).
- While no statistically significant difference was observed between pre- and post-application urinary IMP concentrations in the PET study, the time-concentration profile clearly shows an observable decay in children's urinary biomarker concentrations in the eight days following the outdoor lawn application (Figure 8.7). The pattern among adults is not consistent with that among children.
- Comparing first morning voids (FMV) to other spot samples collected among a subsample of CTEPP children (data not presented), the median concentration in FMV is substantially (43%) higher than the median of the non-FMV samples for TCPy, and slightly (35%) higher for 3-PBA, due to longer urine accumulation time in the bladder.
- In CHAMACOS, concentrations in overnight diapers were compared to concentrations in spot samples (Bradman *et al.*, 2006; data not presented). In all cases, diethyl phosphates were lower in overnight diaper samples than in spot samples, while for toddlers dimethyl phosphates were higher in overnight diaper samples. Median total DAP concentrations for all children were higher in the overnight samples compared to the spot samples (140 *vs.* 100 nmol/l), but the differences were not statistically significant (Wilcoxon test).

- Spearman correlations were calculated for CHAMACOS spot and overnight samples by age (Bradman *et al.*, 2006). Spot and overnight urine concentrations were significantly correlated in CHAMACOS (Bradman *et al.*, 2006): dimethyl phosphate (Spearman rho=0.53; p=0.02), diethyl phosphate (Spearman rho=0.48; p=0.03), and total DAP metabolites (Spearman rho=0.57; p=0.009).

Table 8.6 Intraclass correlation coefficients (ICC) for logged CTEPP urinary metabolites. [a]

Metabolite	NC Children	OH Children
3-PBA	-- [b]	0.70
TCPy	0.65	0.48

[a] An ICC of 0.80 indicates that a single measurement reliably represents the average of a set of measurements.
[b] -- = no data.

Table 8.7 Between- and within-person geometric standard deviations (GSDs) for logged urinary concentrations from children in the CTEPP study.

Metabolite	Measure	NC Children	OH Children
3-PBA	Between-person GSD	-- [a]	1.5
	Within-person GSD	--	1.2
TCPy	Between-person GSD	1.5	1.5
	Within-person GSD	1.3	1.5

[a] -- = no data.

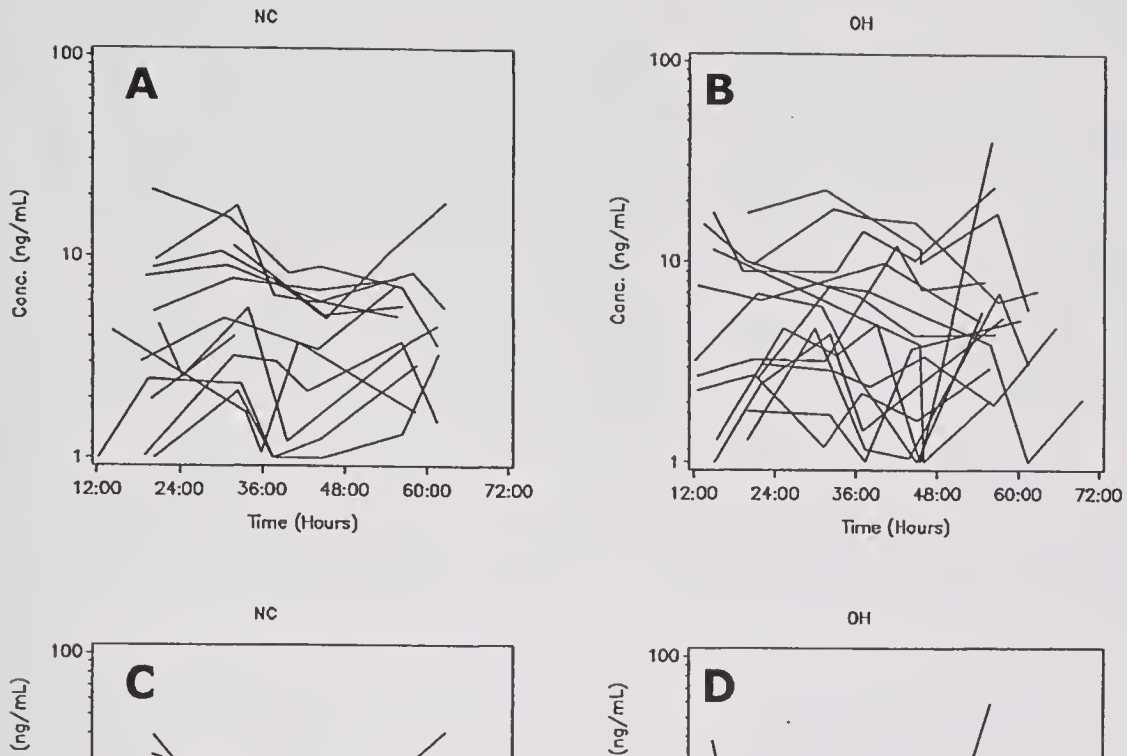


Figure 8.6 Concentration versus time plots for urinary TCPy measurements among CTEPP-NC and CTEPP-OH participants reporting a recent pesticide application. Urines in panels A and B are without adjustment. Urines in panels C and D are adjusted by specific gravity. Note that not all voids within the 48 hour period were collected.

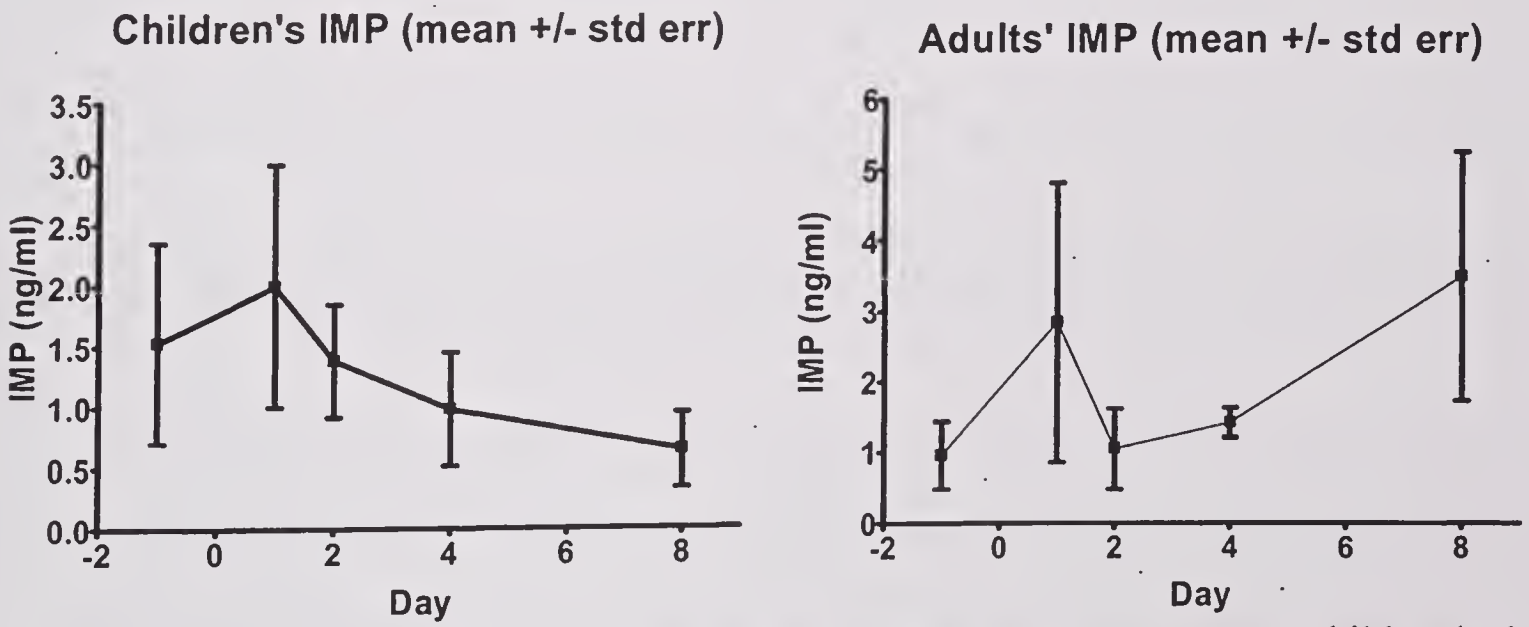


Figure 8.7 Time-concentration profile for urinary IMP measurements among child and adult PET study participants following an outdoor granular turf pesticide application.

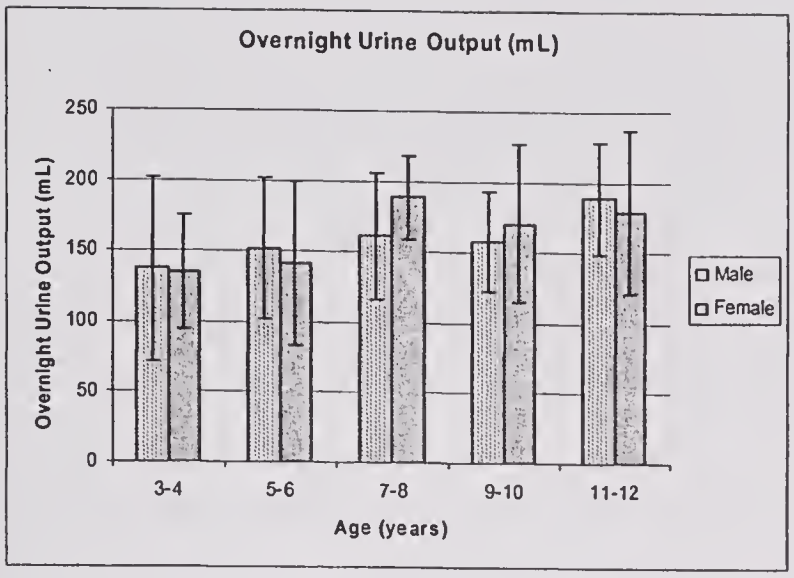
8.4 Urine and Creatinine Excretion among Children

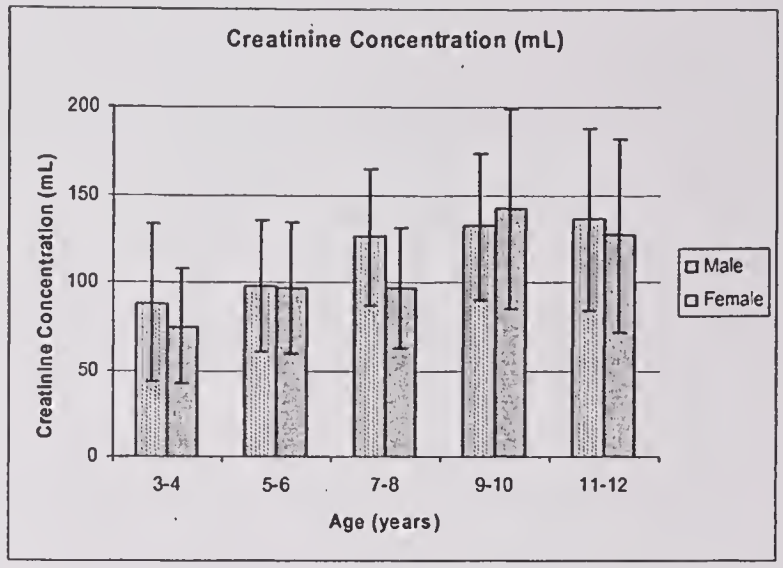
Urine output varies with water intake, urea, salt, specific gravity, and osmolality (Wessels *et al.,* 2003). Consequently, the concentration of metabolites in spot urine samples may vary, even if the internal dose remains constant. Since collecting 24-h urine samples from children is often impractical, spot urine samples are commonly collected and normalized using creatinine (CRE) concentration. However, CRE yield has been shown to be variable among children (Freeman *et al.*, 1995; O'Rourke *et al.*, 2000). Furthermore, because CRE excretion is dependent upon muscle mass, children inherently excrete less CRE than adults. This makes comparisons between CRE-adjusted adult and children urinary biomarker concentrations subject to error due to "over-correction" of children's samples. Age-dependent differences in daily creatinine clearance must also be considered when comparing young children and older ones (Krieger *et al.,* 2001; Wessels et al, 2003), as differences are great even for 1-year olds (0.08 g creatinine/day) relative to 5-year olds (0.4 g creatinine/day).

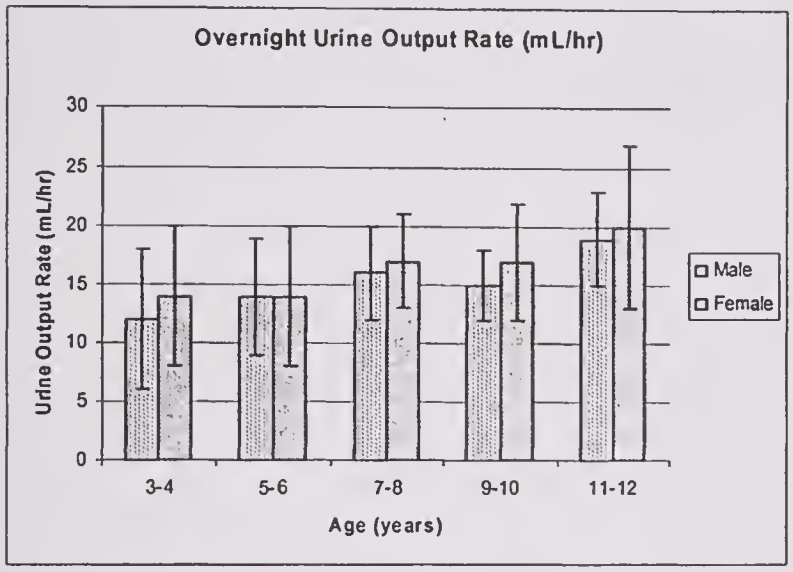
Alternative approaches for adjusting for urine dilution are based on urinary specific gravity and on urinary output. Specific gravity adjustment accounts for all dissolved solids, with a specific gravity of 1.024 considered normal for adults. Both specific gravity and creatinine were measured in CTEPP urine samples.

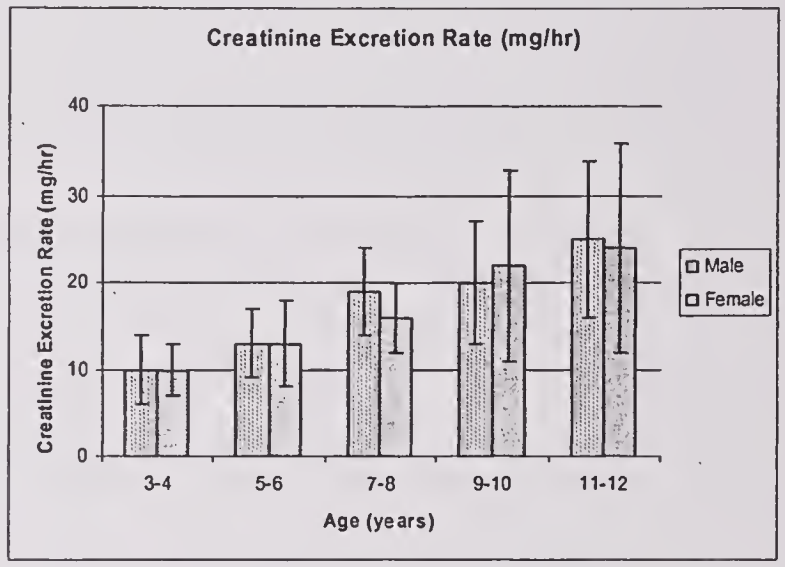
Urinary output among young children is often estimated with equations from the Exposure Factors Handbook. Zartarian et al (2000) estimated daily urinary output volumes of 500 and 800 mL for the children 0–4 and 5–9 years of age, respectively, based on Geigy Scientific Tables. Estimated daily urinary output and creatinine excretion for children 3-12 years of age based on first morning void measurements and recorded ancillary information from the MNCPES are presented in Figure 8.8.

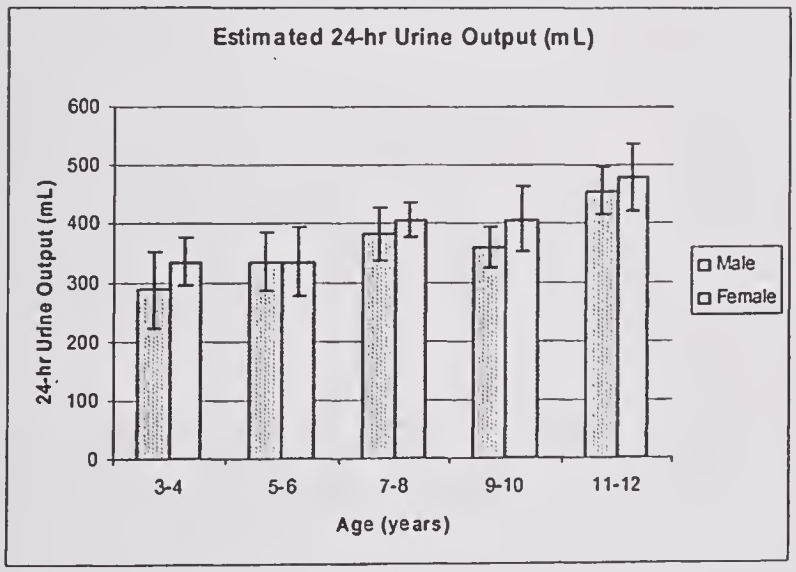
- In unpooled samples from CTEPP, specific gravity of children's urine averaged 1.020, significantly different than the 1.024 of adult urine (t-test, $p < 0.001$).
- In the MNCPES study, the daily urine output rates (mean ± SD) increased from 13 ± 6 mL/hr for 3-4 year olds to 19 ± 7 mL/hr for 11-12 year olds (Figure 8.8) based on first morning void samples with known volumes and void times.
- In the MNCPES study, creatinine excretion rates (mean ± SD) increased from 10 ± 4 mg/hr for 3-4 year olds up to 24 ± 12 mg/hr for 11-12 year olds (Figure 8.8).
- There was neither a substantial nor consistent difference between sexes for either daily urine output or daily creatinine excretion rate, suggesting that sex is not an important predictor of creatinine excretion for pre-pubescent children (Figure 8.8).
- Failure to appropriately account for creatinine excretion results in "over-correction" of children's samples when making comparisons between CRE-adjusted adult and children urinary metabolite concentrations, making child levels appear higher by comparison.
- An alternate approach for avoiding issues with variable urine volumes is to calculate biomarker excretion rates. This requires collection of complete voids, void volume measurements, and recording previous and final void times.











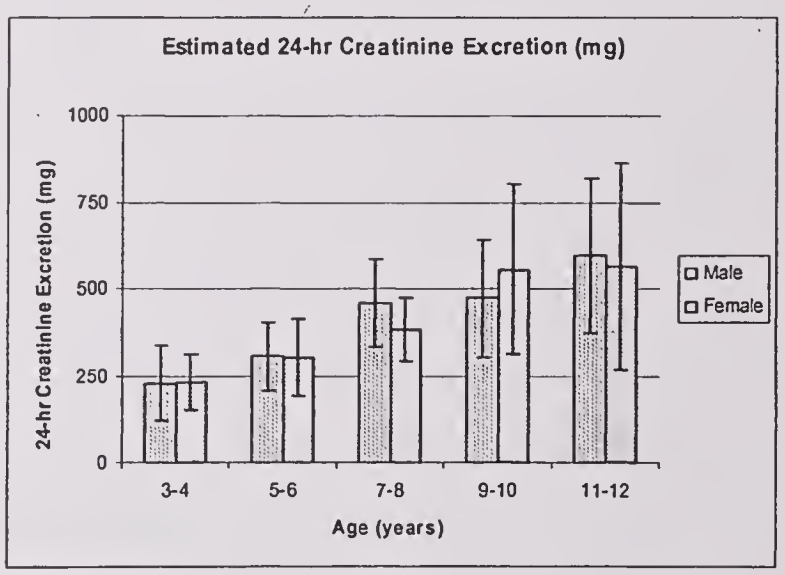


Figure 8.8 Estimates of age-specific urinary output and creatinine excretion, based on data from the MNCPES.

8.5 Relative Importance of Exposure Routes

The relative importance of the dietary ingestion, indirect ingestion, dermal, and inhalation routes of exposure with respect to aggregate intake has been investigated with data from both the MNCPES and CTEPP studies. Daily inhalation and dietary intake estimates (ng/kg/day) for chlorpyrifos among children in MNCPES are available in Clayton *et al.* (2003). Estimated relative importance of the inhalation, dietary ingestion, and indirect ingestion routes of exposure to OPs and pyrethroids among children in CTEPP are presented in Morgan *et al.* (2004).

- MNCPES chlorpyrifos data showed that ingestion was a more dominant route of intake than inhalation. Urinary metabolite levels, however, showed a stronger association with air ($r=0.42$, $p<0.01$) than with dietary ($r=0.22$, $p<0.05$) measurements.
- Using MNCPES data as an input, the SHEDS model suggested (data not presented) that the dominant pathway for highly exposed chlorpyrifos users was non-dietary ingestion, followed by dietary ingestion. The model also suggested that the relative contribution of exposure pathways may differ by pesticide.
- TCPy was found in several environmental media in CTEPP, particularly in solid food samples. Estimated intake of TCPy (Figure 8.9) was about 12 times higher than intake of chlorpyrifos for CTEPP children. Even when environmental TCPy is considered, nearly 60% of the TCPy excreted in urine remained unaccounted for. This suggests that either a major pathway of children's exposure to chlorpyrifos and TCPy remains unaccounted for in our algorithms or that some underlying assumptions are incorrect.
- Despite indications that intake of TCPy from solid food may be responsible for the bulk of TCPy intake, intake from solid food and excretion are poorly correlated ($r^2=0.01$, Figure 8.10). The absorption rate for TCPy remains unknown, as does whether or not it is metabolized to other products in the body.
- Based on exposure algorithms (with absorption assumed to be 50% by each route), the primary route of exposure and intake for chlorpyrifos and permethrin among CTEPP children was dietary ingestion (Table 8.8 and Figure 8.11). Inhalation was the secondary route for chlorpyrifos and diazinon (organophosphates); while indirect ingestion was the secondary route for permethrin (pyrethroid).
- Based on algorithms, the contribution of diet to aggregate intake generally decreases as intake increases (Figure 8.12). Conversely, nondietary ingestion becomes increasingly important with increasing aggregate intake.
- Unlike with TCPy, the estimated aggregate intake of *cis*- and *trans*-permethrin among CTEPP-OH children was close to the excreted amount of 3-PBA (Figure 8.12). However, children may have also been exposed to other pyrethroids that are metabolized into 3-PBA and could have contributed to the excreted amounts measured.
- Our studies consistently report a low correlation between concentrations of urinary biomarkers of pesticide exposure and environmental concentrations. Algorithm-based estimates of aggregate intake do little to improve the correlation. A better understanding of how differences in activities between children affects intake may be needed.
- Figures 8.14 and 8.15 present environmental and dietary levels of chlorpyrifos and

urinary concentrations of TCPy by study. There is little evidence that differences in environmental media concentrations translate into differences in urinary concentrations. The pattern is most similar between food and urine concentrations (Figure 8.15).

Table 8.8 Estimated relative importance of the inhalation, dietary ingestion, and indirect ingestion routes of exposure among children in CTEPP NC and OH.

Class	Pollutants	Apportionment of Aggregated Exposure/Dose
OP Insecticide	Chlorpyrifos and Diazinon	***NC:*** dietary ingestion ≈inhalation > indirect ingestion ***OH:*** dietary ingestion > inhalation > indirect ingestion
Pyrethroid Insecticide	*cis*- and *trans*-Permethrin	***NC:*** dietary ingestion ≈indirect ingestion > inhalation ***OH:*** dietary ingestion > indirect ingestion > inhalation

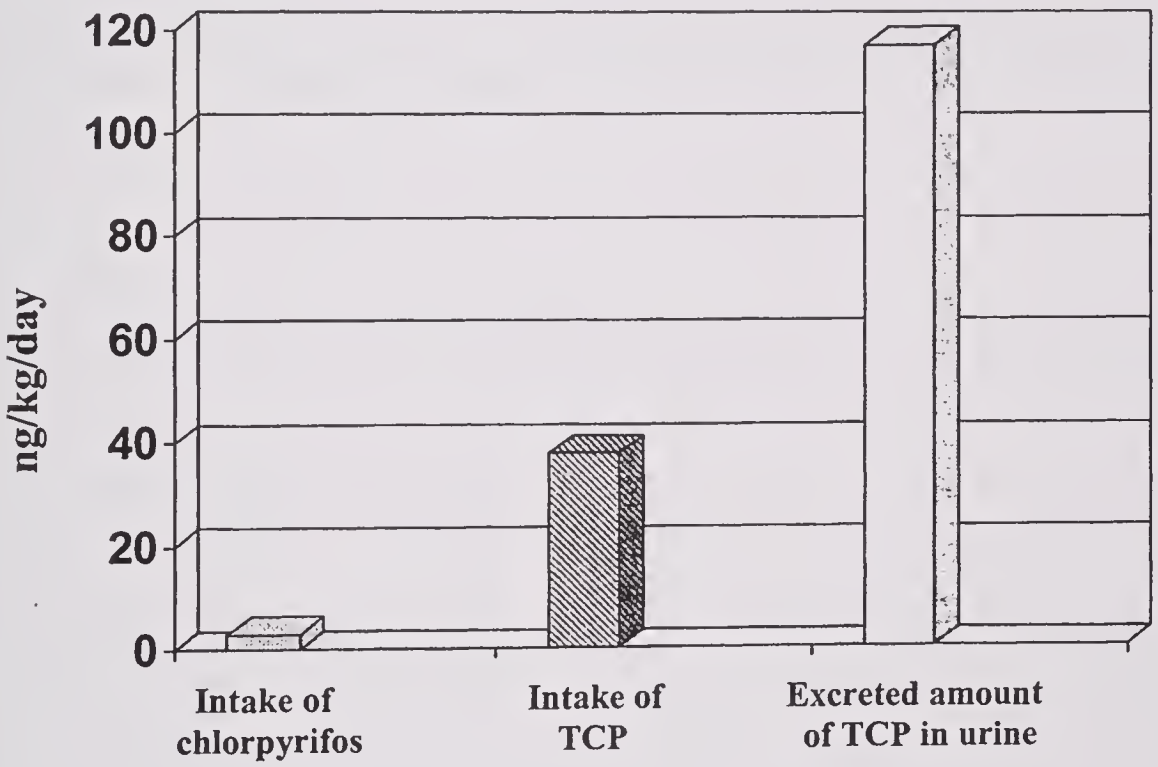


Figure 8.9 The median estimated intakes of chlorpyrifos and TCPy in CTEPP-NC compared with the excreted median amounts of TCPy in the preschool children's urine (Morgan *et al.*, 2005).

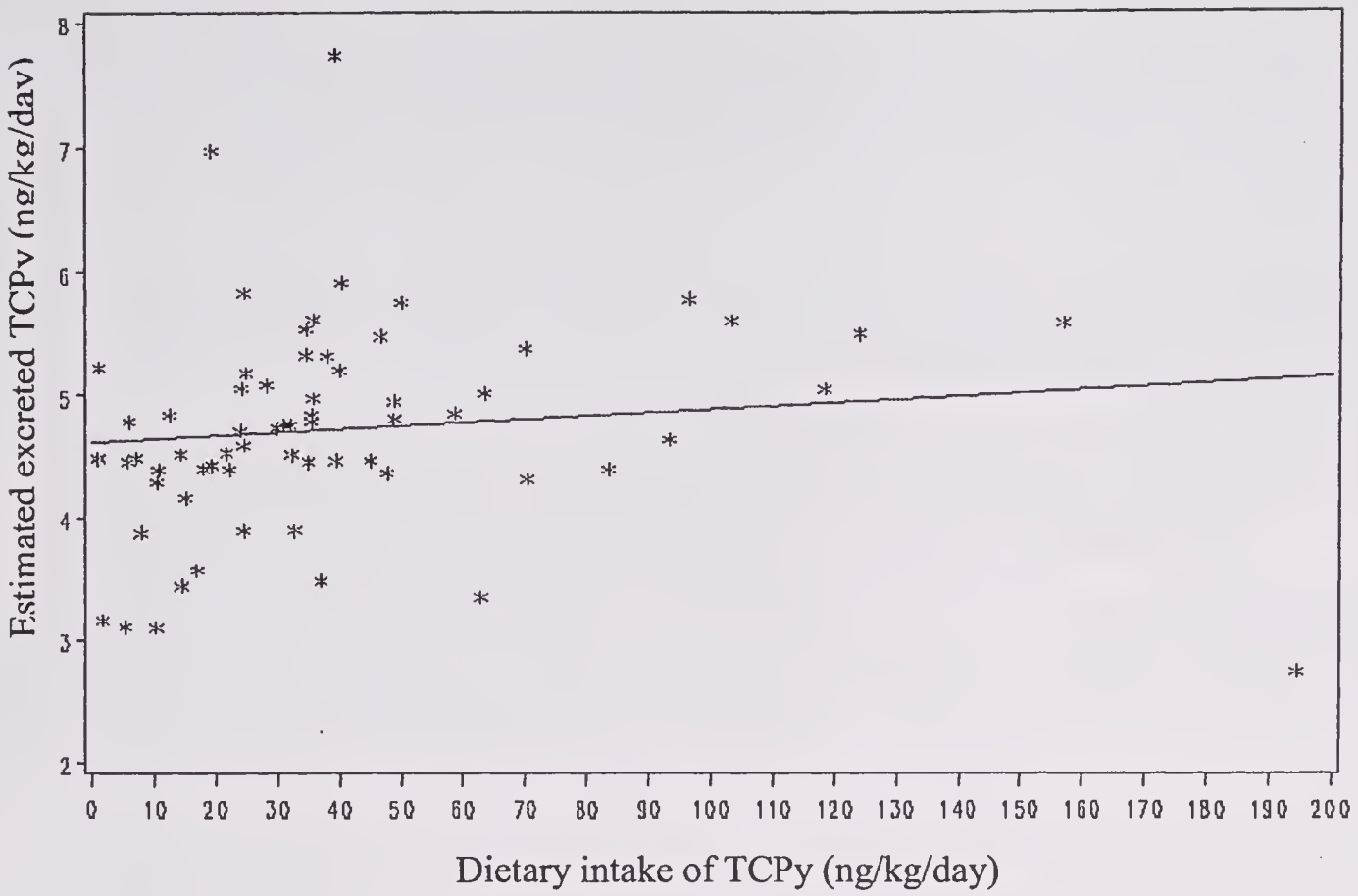


Figure 8.10 Intake of environmental TCPy through the dietary route correlated poorly (r^2=0.01) with the amount of TCPy excreted in the urine of CTEPP-NC preschool children.

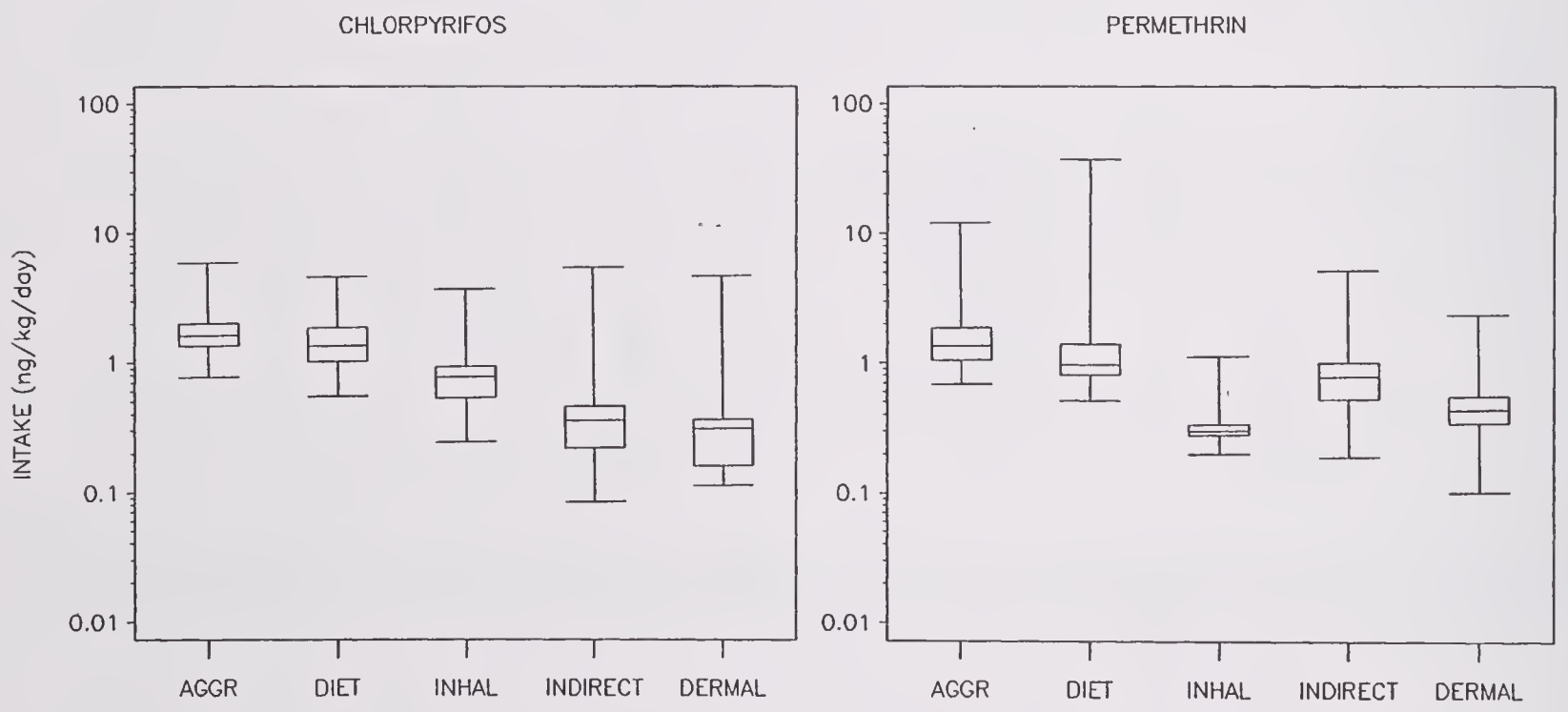


Figure 8.11 Estimated distributions of aggregate intake ("AGGR") of chlorpyrifos and permethrin (ng/kg/day) and estimated distributions of the four contributing routes (diet, inhalation, indirect ingestion, and dermal) among CTEPP-OH children.

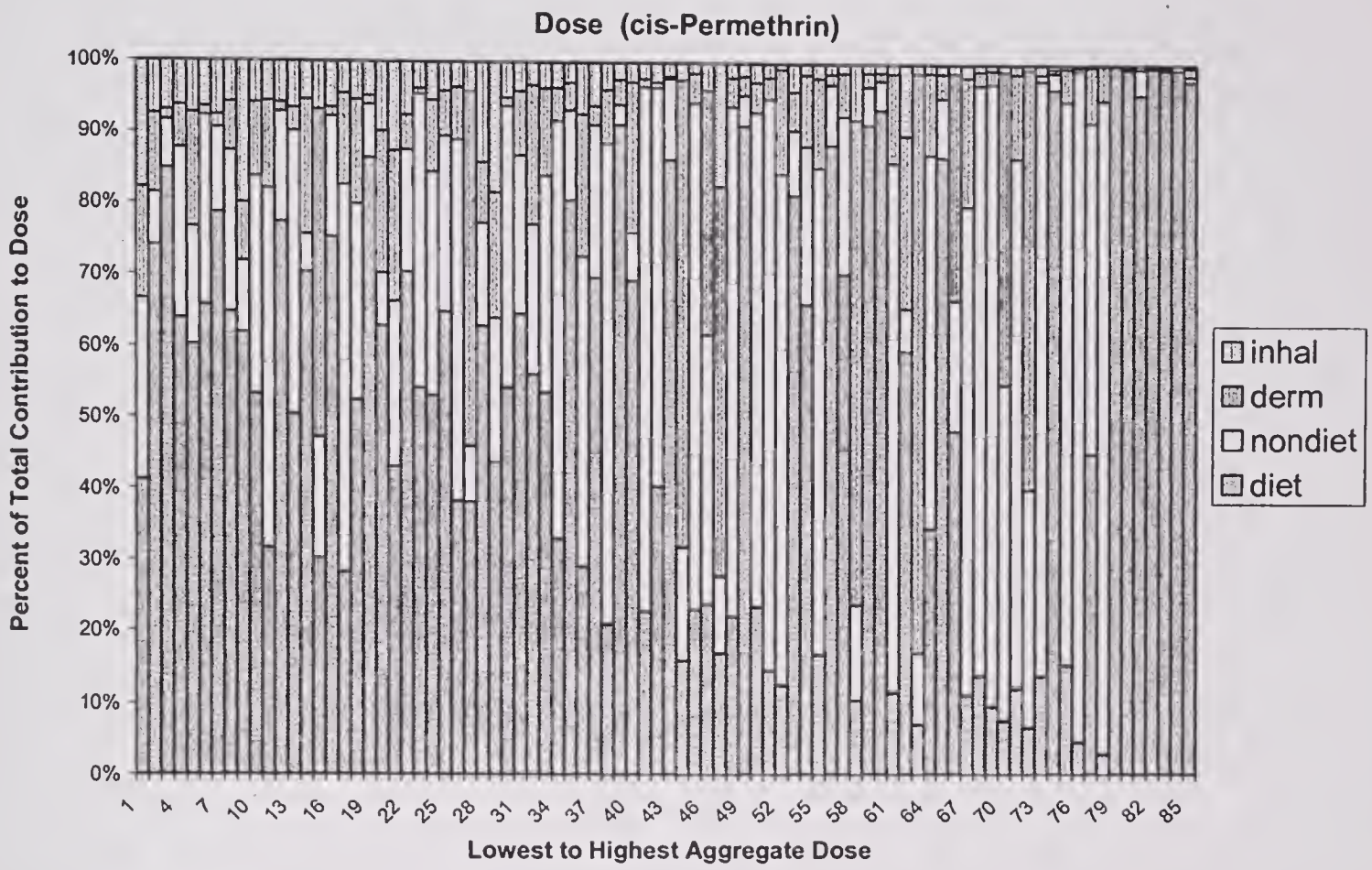


Figure 8.12 The contributions of inhalation, dermal absorption, diet, and nondietary ingestion to aggregate intake of *cis*-permethrin.

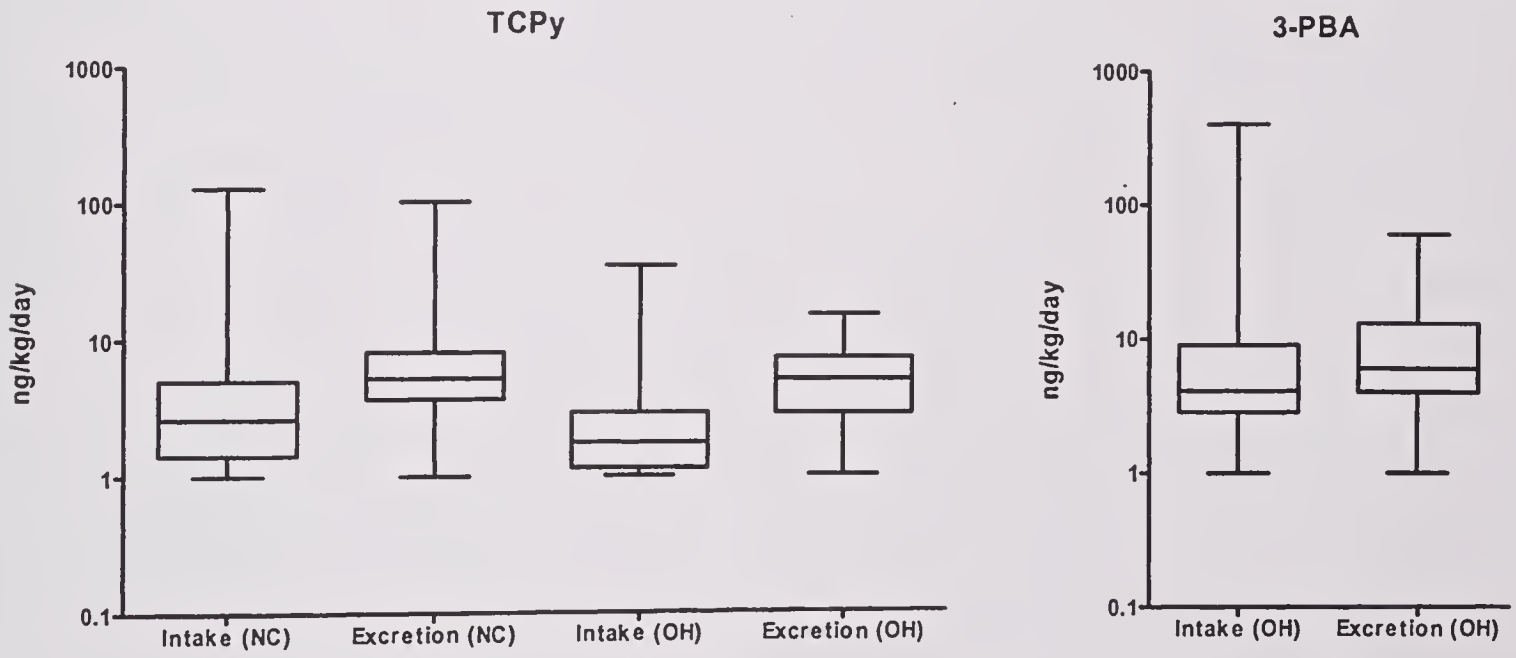


Figure 8.13 Children's estimated aggregate intake of chlorpyrifos and permethrin compared to their measured urinary metabolites (CTEPP).

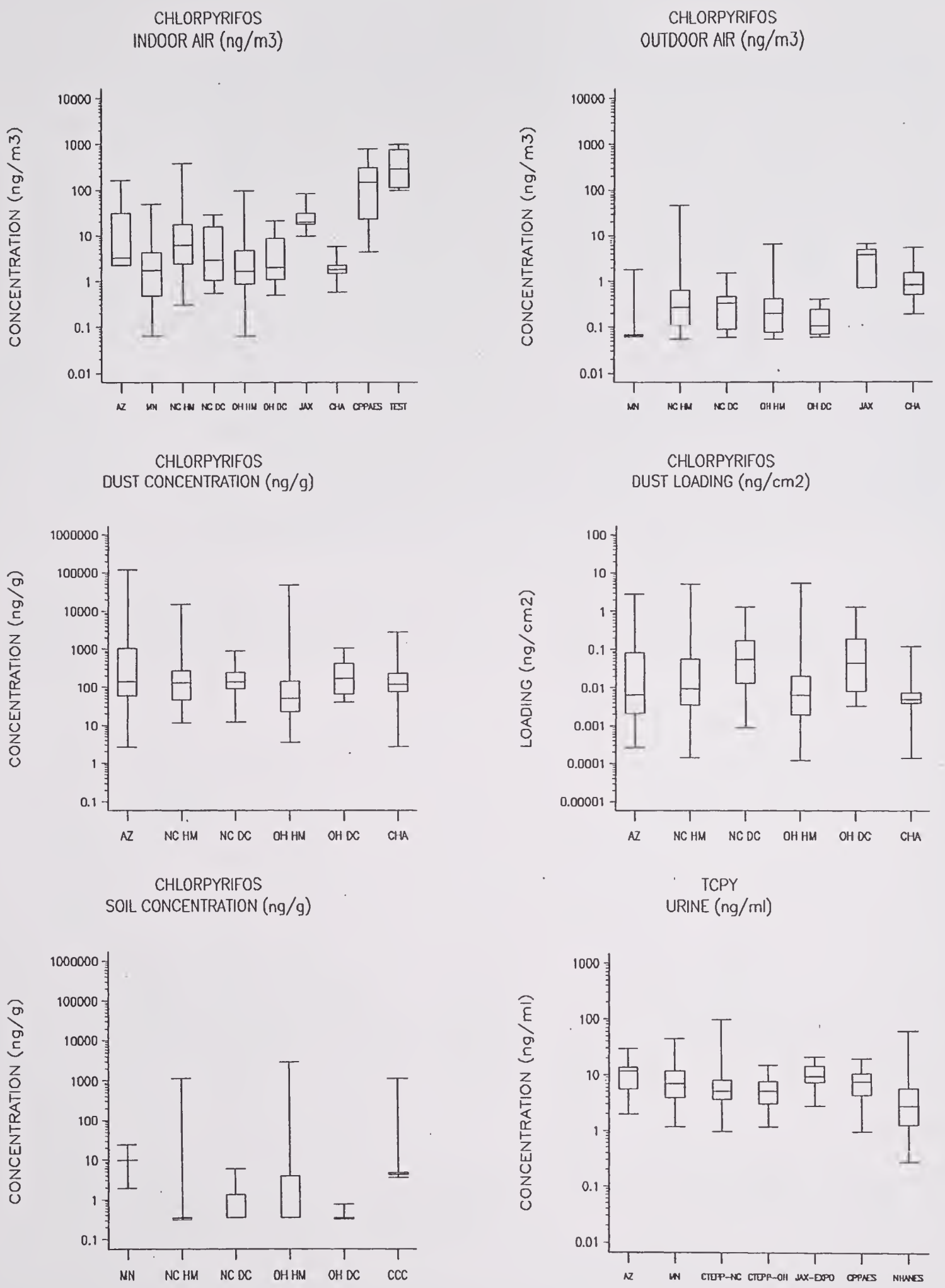


Figure 8.14 Distributions of TCPy in urine across studies (bottom right panel) in comparison to distributions of chlorpyrifos in indoor air, outdoor air, dust, and soil across studies.

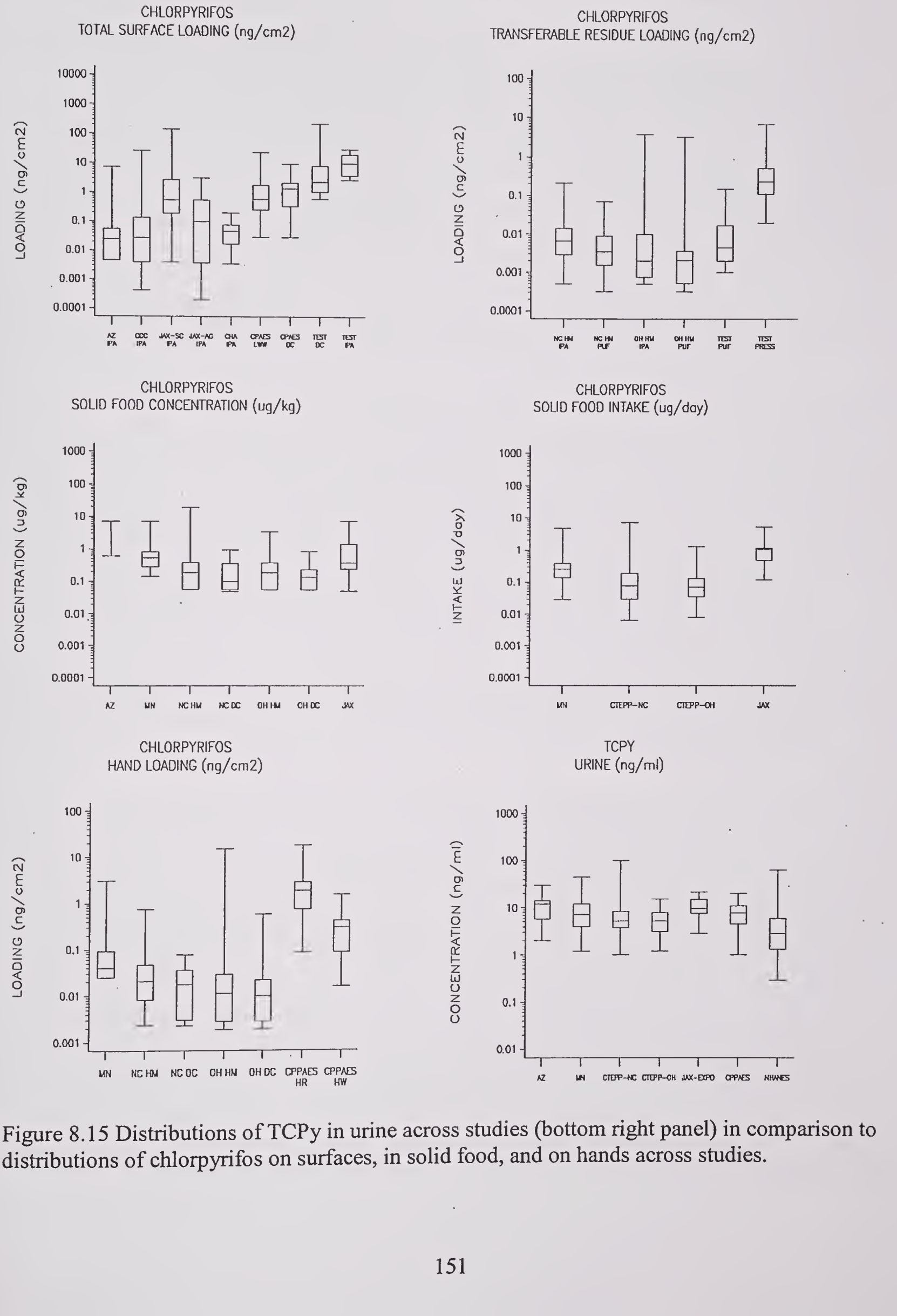


Figure 8.15 Distributions of TCPy in urine across studies (bottom right panel) in comparison to distributions of chlorpyrifos on surfaces, in solid food, and on hands across studies.

8.6 Model Predictions

The Stochastic Human Exposure and Dose Simulation (SHEDS) model (Zartarian *et al.*, 2000) provides route-specific estimates of aggregate exposures, relying on input data from assorted data sets, including those described in this report. The Safe Foods Project is currently developing an exposure-dose-response model to address cumulative risks associated with exposures to multiple pyrethroids. The Project intends to use the Exposure Related Dose Estimating Model (ERDEM) (Blancato *et al.*, 2004) to predict internal dose based on cumulative exposure estimates from SHEDS. The model will be used to identify critical pathways of human exposure and dose. A meaningful discussion of SHEDS and ERDEM is beyond the scope of this report, but an example of an important application of SHEDS is described below.

- Use of the SHEDS model with MNCPES data (Figure 8.16) helped reveal the importance of accounting for exposures to the metabolite/degradate TCPy in environmental media. Without such accounting, the model under-predicted urinary TCPy concentrations.
- SHEDS found that urinary biomarker concentrations depend mainly on dietary intake. An uncertainty analysis (independent of dietary) found other important factors to be: applied pesticide mass; surface area of treated rooms; time in treated rooms; air and residue decay rates; surface-to-skin transfer efficiency; dermal transfer coefficient; saliva removal efficiency; fraction hands mouthed; daily hand wash events; removal efficiency; maximum dermal loading; dermal absorption rate; and frequency of hand-mouth activity.
- By identifying critical pathways of human exposure and dose (and their associated uncertainties), models such as SHEDS and ERDEM guide the planning for future measurement studies so that newly identified data gaps may be filled with real-world measurement data.
- Applying SHEDS to different pesticide classes will provide information on degree to which factors that affect exposure differ across pesticide classes (*e.g.*, pyrethroids *vs.* organophosphates).

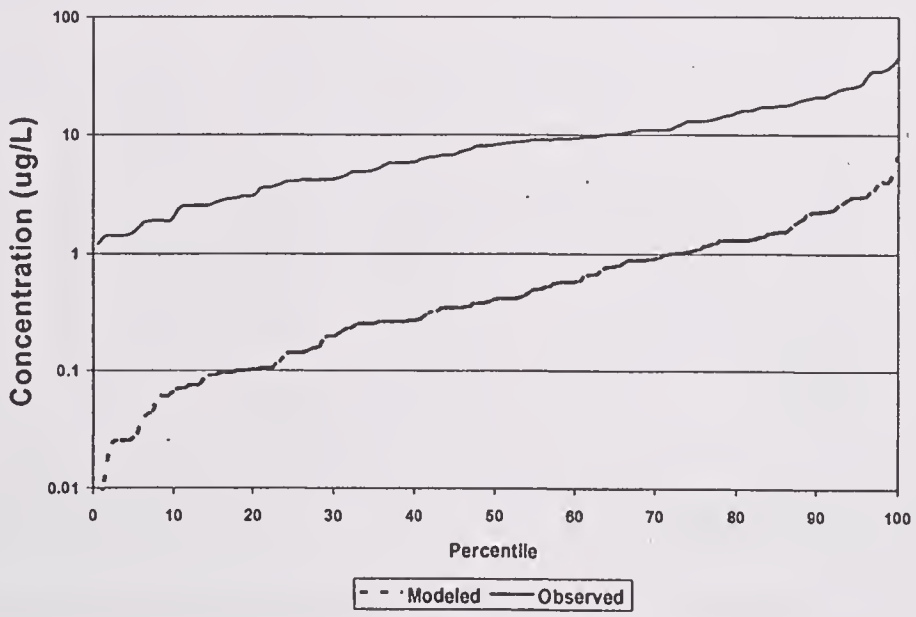


Figure 8.16 Comparison of TCPy in urine between SHEDS model and observed MNCPES data when TCPy in the environment is not considered (Source: Xue *et al.*, 2004).

9.0 SUMMARY AND CONCLUSIONS

In an effort to facilitate risk assessments that take into account unique childhood vulnerabilities to environmental toxicants, the National Exposure Research Laboratory (NERL) in the U.S. Environmental Protection Agency's (U.S. EPA) Office of Research and Development (ORD) identified four priority research areas as representing critical data gaps in our understanding of environmental risks to children. These *priority research areas* are: 1) pesticide use patterns; 2) spatial and temporal distributions of residues in residential dwellings; 3) dermal absorption and indirect (non-dietary) ingestion; and 4) dietary ingestion. Several targeted studies were conducted or financially supported by NERL to specifically address these priority research needs. The studies were designed to address the largest uncertainties associated with children's exposure and aimed to produce sufficient real-world data to eliminate excessive reliance on default assumptions when assessing exposure. Significant progress has been made in each of the four priority areas leading to a more comprehensive understanding of the exposures resulting from children's interactions with their environment.

In the area of pesticide use patterns, our studies have taught us that pesticide products are likely to be found in nearly 9 out of every 10 homes. The most frequently applied of these products typically contain pyrethrins and pyrethroids (namely, permethrin, cypermethrin, and allethrin). The applications are more likely to be performed by an occupant than by a professional, with "crack-and-crevice" type applications favored over either the broadcast or total release aerosol types. The application frequencies appear to be higher in warmer climates, but no differences based on population density (urban *vs.* rural) or other socio-demographic factors including race, ethnicity, home type, income, and level of education are evident. Despite much effort in questionnaire development, we have had little success in correlating questionnaire responses with residue measurements. More effort is still needed to improve questionnaires and to ensure uniformity in inventory forms in future studies. Target populations for future studies should be chosen from areas that extend outside the limited geographic regions that have previously been studied to capture divergent use patterns, but previously studied populations should also be included to document trends.

We have learned a great deal about spatial and temporal distributions of pesticide residues. Indoor air concentrations are typically ten-fold higher than outdoor concentrations, but surprisingly high outdoor air concentrations have also been measured. In the absence of any recent application, concentrations in indoor air are strongly influenced by vapor pressure. Immediately following an application, airborne concentrations peak within 24 hours and produce a concentration gradient with levels decreasing with distance from the application site. Southern states do have higher airborne concentrations than Northern states, but there is considerable overlap. Population density (urban *vs.* rural) and income level differences are evident. With surface residues, considerable variability exists not only among rooms but also in different locations within a room. Substantial translocation of pesticides from application surfaces to adjacent surfaces, and from outdoor surfaces to indoor surfaces has been observed. Cleaning activities and ventilation have been found to be important for both air and surface concentrations. Much, though not all, of what we have learned about spatial and temporal variability has come from organophosphate pesticides, and more studies with pyrethroids are needed.

These studies have added merit to earlier hypotheses that dermal transfer and indirect ingestion are important routes of children's exposure to pesticides. In fact, the shift to less volatile, more organophilic pyrethroid pesticides magnifies the importance of particle-bound transfer and implies an increased significance of indirect ingestion. Substantial challenges still exist in this area. One challenge is to incorporate into estimates of dermal exposure what we have learned through laboratory studies of the importance of skin condition, contact motion, and number of contacts. Another challenge is to standardize the collection methods used to measure the surface residues that are a key part of dermal exposure estimates. A third challenge is to improve our indirect ingestion exposure algorithms to ensure that we are not missing major transfer mechanisms that may bridge the gap between what we are estimating as intake and what we are measuring as excreted.

Analysis of the dietary ingestion components of our studies produce intake estimates that suggest dietary ingestion may often be the dominant route of exposure (even with pyrethroids despite the increased importance of the dermal and indirect ingestion routes). Low detection frequencies in food measurements, however, increase uncertainty, as does the questionable reliability of duplicate diet estimates for young children. Improvements are still essential in both the sample collection and the chemical analysis methods. Large differences in dietary exposure estimates among children in the same studies point to a need for a better understanding of the variability in dietary exposure.

Clearly, more information is needed to assess the relative importance of the exposure routes under different conditions and with pesticides from diverse compound classes. More work is necessary to reconcile aggregate exposure estimates with levels of biomarkers measured in urine. Moreover, more work is needed to better understand how exposures and important exposure factors differ across age groups, as children move through different developmental stages.

We anticipate that the analyses presented in this report will be useful to the EPA Program Offices, including the Office of Pesticide Programs and the Office of Children's Health Protection, in their risk assessment and management activities. Although much of this high-quality, real-world data has already been made available to the Program Offices piecemeal and by publication in the peer reviewed literature, we expect consideration of the data collectively to provide added value to the results of individual studies. Admittedly there are limitations inherent in the comparisons: studies were performed in different seasons, in different years, using different methods, and with different sample sizes. We are confident, however, that these analyses will facilitate more accurate exposure and risk assessments, thereby strengthening regulatory actions aimed at reducing risk, and helping to ensure that pesticides are appropriately regulated.

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APPENDIX A: Summary Statistics

Air Concentrations

Table A.1 Summary statistics for airborne chlorpyrifos concentrations (ng/m³) by study.

Study	Location	N	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
NHEXAS-AZ	Indoor	14	50	25.9	44.6	8.13	4.7	<3.2	<3.2	3.37	31.6	165	165
	Outdoor	3	0	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC
MNCPES	Personal	61	95	6.05	17.6	1.91	4.2	<0.10	0.93	1.52	4.61	16.9	135
	Indoor	80	93	5.61	10.1	1.71	5.1	<0.10	0.50	1.85	4.40	30.3	49.5
	Outdoor	52	6	0.09	NC	NC	NC	<0.10	<0.10	<0.10	<0.10	0.19	0.91
CTEPP-NC	Indoor	148	100	17.5	39.3	6.45	4.0	0.31	2.26	6.07	17.3	62.2	391
	Outdoor	140	83	1.00	4.02	0.30	3.6	<0.10	0.11	0.28	0.64	3.99	45.9
CTEPP-OH	Indoor	147	98	6.24	13.8	2.26	3.7	<0.10	0.93	1.75	5.82	21.7	98.0
	Outdoor	126	75	0.39	0.75	0.21	2.7	<0.10	0.07	0.20	0.39	1.13	6.50
JAX	Indoor	9	100	30.0	23.3	24.3	1.9	9.81	18.3	20.4	32.4	84.9	84.9
	Outdoor	9	56	3.05	2.35	2.06	2.8	<1.0	<1.0	3.77	4.94	6.62	6.62
CHAMACOS	Indoor	20	100	2.0	1.1	1.8	1.6	0.6	1.5	1.9	2.3	4.4	5.9
	Outdoor	19	84	1.2	1.1	0.8	2.3	<0.3	0.5	0.90	1.5	5.5	5.5
CPPAES (Day 1)	Indoor	10	100	204	247	86.3	5.1	4.55	23.9	150	312	816	816
Test House (Day 1)	Indoor	6	100	431	376	301	2.6	100	115	290	790	1000	1000

NC, not calculated due to low detection frequency

Table A.2 Summary statistics for airborne diazinon concentrations (ng/m³) by study.

Study	Location	N	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
NHEXAS –AZ	Indoor	14	64	30.9	61.4	7.22	5.4	<2.0	<2.0	5.59	12.0	220	220
	Outdoor	3	0	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC
MNCPES	Personal	48	65	1.88	7.86	0.34	4.5	<0.10	<0.10	0.28	0.82	4.66	54.5
	Indoor	73	66	1.68	5.76	0.35	4.7	<0.10	<0.10	0.27	0.81	8.59	47.1
	Outdoor	52	12	0.29	NC	NC	NC	<0.10	<0.10	<0.10	<0.10	0.22	10.2
CTEPP-NC	Indoor	148	100	36.4	202	2.42	6.0	0.14	0.66	2.03	5.09	63.7	1780
	Outdoor	140	52	0.59	3.70	0.13	3.0	<0.10	<0.10	0.09	0.22	0.98	42.8
CTEPP-OH	Indoor	147	98	11.8	48.0	1.41	5.3	<0.10	0.51	0.97	2.41	56.9	482
	Outdoor	143	74	1.09	6.91	0.19	3.3	<0.10	<0.10	0.15	0.33	1.49	78.9
JAX	Indoor	9	78	7.18	8.45	3.43	4.7	<0.40	3.43	4.64	8.05	28.0	28.0
	Outdoor	9	67	3.45	2.63	1.89	4.2	<0.40	<0.40	3.53	5.78	6.76	6.76
CHAMACOS	Indoor	20	100	5.2	9.8	2.5	2.8	1.0	1.3	1.8	2.8	29	44
	Outdoor	19	100	5.3	6.1	3.3	2.6	1.0	1.4	2.8	5.3	21	21
DIYC	Indoor	16	100	2280	1790	1470	3.0	245	541	1840	4060	4900	4900
PET	Indoor	60	77	127	196	25.8	10.7	<0.85	7.60	45.6	163	562	1040

NC, not calculated due to low detection frequency

Table A.3 Summary statistics for airborne malathion concentrations (ng/m³) by study.

Study	Location	N	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
NHEXAS-AZ	Indoor	14	14	NC	NC	NC	NC	<3.0	<3.0	<3.0	<3.0	5.61	5.61
	Outdoor	3	33	NC	NC	NC	NC	<3.0	<3.0	<3.0	6.85	6.85	6.85
MNCPES	Indoor	88	67	1.53	1.87	0.59	5.3	<0.10	<0.10	1.18	2.11	4.82	13.0
	Outdoor	51	12	NC	NC	NC	NC	<0.10	<0.10	<0.10	<0.10	0.76	1.95
JAX	Indoor	9	0	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC
	Outdoor	9	11	NC	NC	NC	<1.4	<1.4	<1.4	<1.4	<1.4	6.57	6.57
CHAMACOS	Indoor	20	15	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	5.6	7.8
	Outdoor	19	37	NC	NC	NC	NC	<0.5	<0.5	<0.5	2.6	17	17

NC, not calculated due to low detection frequency

Table A.4 Summary statistics for airborne *cis*-permethrin concentrations (ng/m³) by study.

Study	Location	N	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	Personal	64	86	0.78	2.21	0.23	4.1	<0.09	0.09	0.20	0.61	2.07	15.7
	Indoor	89	69	0.53	2.34	0.11	3.8	<0.09	<0.09	0.09	0.18	1.26	20.9
	Outdoor	51	43	NC	NC	NC	NC	<0.04	<0.04	<0.04	0.06	0.15	0.23
CTEPP-NC	Indoor	148	65	1.91	4.83	0.42	5.5	<0.10	<0.10	0.41	1.43	7.79	34.4
	Outdoor	140	19	NC	NC	NC	NC	<0.10	<0.10	<0.10	<0.10	0.47	1.62
CTEPP-OH	Indoor	147	22	NC	NC	NC	NC	<0.40	<0.40	<0.40	<0.40	1.63	6.50
	Outdoor	143	21	NC	NC	NC	NC	<0.40	<0.40	<0.40	<0.40	0.95	1.78
JAX	Indoor	9	44	NC	NC	NC	NC	<1.0	<1.0	<1.0	2.21	92.5	92.5
	Outdoor	9	56	1.55	0.80	1.34	1.8	<1.0	<1.0	2.13	2.22	2.29	2.29
CHAMACOS	Indoor	20	40	NC	NC	NC	NC	<0.6	<0.6	<0.6	0.77	1.2	1.3
	Outdoor	20	32	NC	NC	NC	NC	<0.6	<0.6	<0.6	1.1	1.4	1.4

NC, not calculated due to low detection frequency

Table A.5 Summary statistics for airborne *trans*-permethrin concentrations (ng/m³) by study.

Study	Location	N	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	Personal	68	63	0.61	1.95	0.11	5.3	<0.09	<0.09	<0.09	0.38	1.72	13.9
	Indoor	96	42	NC	NC	NC	NC	<0.09	<0.09	<0.09	0.09	1.26	18.0
	Outdoor	51	14	NC	NC	NC	NC	<0.09	<0.09	<0.09	<0.09	0.48	8.12
CTEPP-NC	Indoor	148	63	1.72	4.89	0.35	5.3	<0.10	<0.10	0.27	1.16	7.16	40.9
	Outdoor	140	19	NC	NC	NC	NC	<0.10	<0.10	<0.10	<0.10	0.30	1.01
CTEPP-OH	Indoor	147	19	NC	NC	NC	NC	<0.40	<0.40	<0.40	<0.40	1.04	6.84
	Outdoor	143	17	NC	NC	NC	NC	<0.40	<0.40	<0.40	<0.40	0.66	1.32
JAX	Indoor	9	67	17.8	43.7	3.49	5.3	<1.0	<1.0	3.06	6.38	134	134
	Outdoor	9	78	3.51	3.01	2.54	2.4	<1.0	2.08	2.50	4.55	10.2	10.2
CHAMACOS	Indoor	19	16	NC	NC	NC	NC	<0.6	<0.6	<0.6	<0.6	1.8	1.8
	Outdoor	18	0	NC	NC	NC	NC	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6

NC, not calculated due to low detection frequency

Table A.6 Summary statistics for airborne TCPy concentrations (ng/m³) by study.

Study	Location	N	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP-NC	Indoor	148	99	4.68	12.47	1.78	3.8	<0.09	0.81	1.77	3.99	14.3	1040
	Outdoor	140	88	0.44	0.91	0.24	2.6	<0.09	0.13	0.22	0.40	1.57	9.06
CTEPP-OH	Indoor	144	100	1.97	4.62	0.84	3.1	0.09	0.43	0.65	1.74	8.60	42.0
	Outdoor	133	88	0.32	0.48	0.22	2.2	<0.09	0.13	0.21	0.36	0.88	4.86

Table A.7 Summary statistics for airborne IMP concentrations (ng/m³) by study.

Study	Location	N	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP-OH	Indoor	147	95	1.52	3.62	0.64	3.1	<0.09	0.35	0.53	1.04	5.68	27.4
	Outdoor	141	86	1.48	5.93	0.36	3.7	<0.09	0.14	0.33	0.77	2.44	49.6

Dust and Soil Concentrations and Loadings

Table A.8 Summary statistics for chlorpyrifos concentrations measured in soil (ng/g).

. Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	Soil	Home	102	3	NC	NC	NC	NC	<10	<10	<10	<10	<10	24.9
CTEPP (NC)	Soil	Home	128	19	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	16.7	1170
		Daycare	13	8	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	0.76	0.76
CTEPP (OH)	Soil	Home	127	39	NC	NC	NC	NC	<0.5	<0.5	<0.5	3.92	13.8	2930
		Daycare	16	38	NC	NC	NC	NC	<0.5	<0.5	<0.5	1.32	6.16	6.16
CCC	Soil	Daycare	117	23	NC	NC	NC	NC	<5	<5	<5	<5	26.8	1150

NC, Not calculated

Table A.9 Summary statistics for chlorpyrifos measured in dust, presented as both loading (ng/cm^2) and concentration (ng/g).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Loading (ng/cm^2)	NHEXAS-AZ	Vacuum	Children ≤12	13	77	0.34	0.80	0.012	23	<0.002	0.002	0.007	0.086	2.81	2.81
	CTEPP (NC)	Floor Dust	Home	121	100	0.14	0.63	0.140	7.3	0.0001	0.0034	0.0094	0.056	0.42	5.16
			Daycare	19	100	0.21	0.37	0.055	6.4	0.0009	0.014	0.057	0.18	1.32	1.32
	CTEPP (OH)	Floor Dust	Home	120	100	0.106	0.54	0.008	6.9	0.0001	0.002	0.006	0.02	0.35	5.41
			Daycare	23	100	0.19	0.33	0.044	6.8	0.003	0.008	0.045	0.19	0.89	1.34
	CHAMACOS	House Dust	All	20	95	0.014	0.030	0.005	4.2	<0.001	0.004	0.005	0.008	0.098	0.12
Concentration (ng/g)	CTEPP (NC)	Floor Dust	Home	121	100	413	1430	137	3.7	11.5	47.5	135	281	1180	15100
			Daycare	19	100	237	256	132	3.5	12.4	94.2	142	254	921	921
	CTEPP (OH)	Floor Dust	Home	120	100	871	5030	70.4	5.1	3.62	23.1	52.0	149	1410	49600
			Daycare	23	100	272	285	168	2.7	40.6	67.0	174	430	897	1110
	CHAMACOS	House Dust	All	20	95	370	684	128	4.7	<4	78.5	120	242	2180	2840

Table A.10 Summary statistics for diazinon concentrations measured in soil (ng/g).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	Soil	Home	102	4	NC	NC	NC	NC	<10	<10	<10	<10	<10	24.9
CTEPP (NC)	Soil	Home	129	18	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	4.24	5470
		Daycare	13	0	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
CTEPP (OH)	Soil	Home	127	34	NC	NC	NC	NC	<0.5	<0.5	<0.5	0.99	4.72	28500
		Daycare	16	19	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	7.07	7.07
CCC	Soil	Daycare	117	20	NC	NC	NC	NC	<2	<2	<2	<2	21.9	110000
PET	Soil	Home	4	100	16900	6140	16000	1.45	10100	12600	16200	21100	24900	2490

NC, Not calculated

Table A.11 Summary statistics for diazinon measured in dust, presented as both loading (ng/cm²) and concentration (ng/g).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Loading (ng/cm²)	NHEXAS-AZ	Vacuum	Children ≤12	13	54	0.035	0.062	0.007	7.1	<0.002	<0.002	0.002	0.035	0.18	0.18
	CTEPP (NC)	Floor Dust	Home	121	96	0.0964	0.638	0.0025	8.8	<0.0003	0.0006	0.0016	0.0106	0.123	5.63
			Daycare	19	100	0.571	2.25	0.0235	11	0.0002	0.0032	0.0177	0.154	9.86	9.86
	CTEPP (OH)	Floor Dust	Home	9120	96	0.094	0.59	0.004	7.5	<0.0003	0.001	0.002	0.01	0.31	6.24
			Daycare	23	100	0.1	0.27	0.02	5.9	0.001	0.004	0.022	0.06	0.39	1.25
	CHAMACOS	House Dust	All	20	100	0.0065	0.018	0.0022	3.2	0.0004	0.0010	0.0021	0.0032	0.048	0.081
	PET	Floor Dust	All	17	100	5.72	16.5	0.44	2.4	0.005	0.092	0.35	1.4	68	68
Concentration (ng/g)	CTEPP (NC)	Floor Dust	Home	121	96	282	1380	24.4	5.1	<2	7.90	17.5	54.4	388	11000
			Daycare	19	100	439	1560	58.6	5.6	3.06	26.0	65.2	138	6880	6880
	CTEPP (OH)	Floor Dust	Home	120	96	1360	8470	34.3	7.2	<2	9.72	19.8	73.2	1710	79900
			Daycare	23	100	260	472	73.7	4.8	5.08	28.4	40.0	210	1610	1630
	CHAMACOS	House Dust	All	20	100	202	562	53.9	3.9	7.75	21.3	58.8	74.4	1470	2550
	PET	Floor Dust	All	17	100	29200	53000	4990	2.1	256	654	312	18500	149000	149000

NC, Not calculated

Table A.12 Summary statistics for *cis*-permethrin concentrations measured in soil (ng/g).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	Soil	Home	102	3	NC	NC	NC	NC	<10	<10	<10	<10	<10	24.9
CTEPP (NC)	Soil	Home	128	19	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	16.7	1170
		Daycare	13	8	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	0.76	0.76
CTEPP (OH)	Soil	Home	127	39	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	13.8	2930
		Daycare	16	0	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
CCC	Soil	Daycare	117	23	NC	NC	NC	NC	<5	<5	<5	<5	26.8	1150

NC, Not calculated

Table A.13 Summary statistics for *cis*-permethrin measured in dust, presented as both loading (ng/cm^2) and concentration (ng/g).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Loading (ng/cm^2)	CTEPP (NC)	Floor Dust	Home	121	100	0.975	3.02	0.104	8.8	0.0012	0.026	0.103	0.411	4.94	23.0
			Daycare	20	100	5.44	19.6	0.507	8.3	0.005	0.181	0.694	1.78	46.9	88.3
	CTEPP (OH)	Floor Dust	Home	120	100	0.83	4.32	0.063	7.5	0.002	0.015	0.045	0.25	3.85	45.4
			Daycare	23	100	0.78	1.36	0.26	5.0	0.01	0.07	0.27	0.68	4.82	5.03
	CHAMACOS	House Dust	All	20	100	0.030	0.063	0.013	3.4	0.0013	0.0057	0.015	0.021	0.17	0.29
Concentration (ng/g)	CTEPP (NC)	Floor Dust	Home	121	100	6080	29400	995	4.6	67.1	347	804	1850	21100	311000
			Daycare	20	100	3500	6760	1140	4.3	113	455	806	2230	19700	29000
	CTEPP (OH)	Floor Dust	Home	120	100	2320	8050	572	4.3	16.6	197	470	1550	7630	79600
			Daycare	23	100	1460	1300	968	2.6	127	418	1010	1850	3830	4630
	CHAMACOS	House Dust	All	20	100	923	2010	317	4.2	25.6	113	345	598	5810	9070

Table A.14 Summary statistics for *trans*-permethrin concentrations measured in soil (ng/g).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Soil	Home	129	22	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	17.9	1610
		Daycare	13	8	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	2.20	2.20
CTEPP (OH)	Soil	Home	124	6	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	2.06	1400
		Daycare	14	0	NC	NC	NC	NC	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
CCC	Soil	Daycare	117	16	NC	NC	NC	NC	<5	<5	<5	<5	12.0	136

NC, Not calculated

Table A.15 Summary statistics for *trans*-permethrin measured in dust, presented as both loading (ng/cm²) and concentration (ng/g).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Loading (ng/cm²)	CTEPP (NC)	Floor Dust	Home	121	100	0.94	2.99	0.09	10	0.0006	0.015	0.09	0.38	4.42	22.6
			Daycare	20	100	5.59	20.2	0.49	8.2	0.005	0.137	0.41	1.38	48.8	91.2
	CTEPP (OH)	Floor Dust	Home	118	100	0.76	4.26	0.05	8.2	0.002	0.010	0.03	0.14	3.86	45.0
			Daycare	22	100	0.73	1.40	0.20	6.0	0.007	0.047	0.26	0.57	4.72	5.17
	CHAMACOS	House Dust	All	20	100	0.06	0.13	0.03	3.4	0.002	0.014	0.02	0.06	0.38	0.58
Concentration (ng/g)	CTEPP (NC)	Floor Dust	Home	121	100	6120	30400	835	5.0	51.3	267	629	1850	19400	322000
			Daycare	20	100	3600	7120	1110	4.5	125	542	856	1830	20900	29900
	CTEPP (OH)	Floor Dust	Home	118	100	2340	8320	453	5.0	16.5	132	344	1270	9210	78800
			Daycare	22	100	1260	1220	784	2.7	126	362	554	1860	3420	3950
	CHAMACOS	House Dust	All	20	100	1860	4030	655	4.0	43.2	310	608	1250	11300	18200

Table A.16 Summary statistics for cyfluthrin concentrations measured in soil (ng/g).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Soil	Home	129	12	NC	NC	NC	NC	<5	<5	<5	<5	32.1	187
		Daycare	13	8	NC	NC	NC	NC	<5	<5	<5	<5	42.2	42.2
CTEPP (OH)	Soil	Home	127	17	NC	NC	NC	NC	<5	<5	<5	<5	64.2	644
		Daycare	16	25	NC	NC	NC	NC	<5	<5	<5	<5	42.2	42.2
CCC	Soil	Daycare	117	10	NC	NC	NC	NC	<6	<6	<6	<6	8.58	11000

NC, Not calculated

Table A.17 Summary statistics for cyfluthrin measured in dust, presented as both loading (ng/cm^2) and concentration (ng/g).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Loading (ng/cm^2)	CTEPP (NC)	Floor Dust	Home	121	48	NC	NC	NC	NC	<0.0003	<0.0003	<0.0003	0.04	0.16	2.14
			Daycare	19	42	NC	NC	NC	NC	<0.0003	<0.0003	<0.0003	0.31	0.78	0.78
	CTEPP (OH)	Floor Dust	Home	119	74	0.056	0.10	0.016	5.6	<0.0003	<0.0003	0.018	0.054	0.25	0.66
			Daycare	23	74	0.37	0.5	0.059	14	<0.0003	<0.0003	0.14	0.74	1.1	1.9
	CHAMACOS	House Dust	All	20	10	NC	NC	NC	NC	<0.005	<0.005	<0.005	<0.005	0.027	0.030
Concentration (ng/g)	CTEPP (NC)	Floor Dust	Home	121	48	NC	NC	NC	NC	<10	<10	<10	248	1660	4100
			Daycare	19	42	NC	NC	NC	NC	<10	<10	<10	329	1750	1750
	CTEPP (OH)	Floor Dust	Home	119	74	329	482	148	3.9	<10	<10	195	384	1280	3040
			Daycare	23	74	389	323	221	3.7	<10	<10	336	648	890	1010
	CHAMACOS	House Dust	All	20	10	NC	NC	NC	NC	<100	<100	<100	<100	828	949

NC, Not calculated

Table A.18 Summary statistics for TCPy concentrations measured in soil (ng/g).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Soil	Home	129	71	3.61	14.9	0.62	4.22	<0.2	<0.2	0.57	1.25	10.7	111
		Daycare	13	46	NC	NC	NC	NC	<0.2	<0.2	<0.2	0.35	1.70	1.70
CTEPP (OH)	Soil	Home	127	80	3.99	15.3	0.82	4.35	<0.2	0.23	0.70	2.02	8.86	127
		Daycare	16	81	1.15	1.57	0.60	3.17	<0.2	0.22	0.63	1.35	6.30	6.30

NC, Not calculated

Table A.19 Summary statistics for IMP concentrations measured in soil (ng/g).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (OH)	Soil	Home	125	41	NC	NC	NC	NC	<0.2	<0.2	<0.2	0.43	2.07	162
		Daycare	16	38	NC	NC	NC	NC	<0.2	<0.2	<0.2	0.44	1.43	1.43

NC, Not calculated

Total Available Surface Residue Loadings

Table A.20 Summary statistics for chlorpyrifos in Total Available Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
NHEXAS-AZ	Surface Wipe	Window Sill	6	17	NC	NC	NC	NC	<0.07	<0.07	<0.07	<0.07	7.49	7.49
MNCPES	LWW	Floor	99	62	1.04	0.41	0.83	1.4	<1.15	<1.15	<1.15	1.15	1.51	3.64
CCC	Surface Wipe	Floor	168	64	0.38	2.28	0.027	7.7	<MDL	<MDL	0.02	0.13	0.97	27.58
		Desk/Table	80	73	0.18	0.53	0.036	6.4	<MDL	0.004	0.04	0.13	0.67	4.29
JAX	Surface Wipe	Floor (Screening)	46	87	4.87	20.32	0.44	12.5	<MDL	0.16	0.50	2.71	10.22	138.4
		Floor	9	78	0.85	1.11	0.21	12.0	<MDL	0.16	0.39	0.72	3.12	3.12
		Play Area	9	67	0.32	0.77	0.014	17.0	<MDL	<MDL	0.006	0.04	2.33	2.33
CHAMACOS	Surface Wipe	All	20	95	0.060	0.057	0.037	2.96	<MDL	0.017	0.046	0.079	0.19	0.20
CPPAES	LWW	Living Area/Kitchen (Pre-application)	20	60	0.29	0.38	0.1	4.91	0.02	0.02	0.099	0.57	1.04	1.22
		Living Area/Kitchen	97	100	2.39	4.30	0.95	3.68	0.07	0.43	0.82	1.96	10.85	24.64
		Bedroom (Pre-application)	20	65	0.41	0.48	0.16	5.24	0.02	0.02	0.26	0.61	1.57	1.90
		Bedroom	64	100	1.97	4.84	0.52	4.40	0.031	0.18	0.35	1.42	6.57	23.76
	Deposition Coupons	Cumulative	39	100	2.12	2.66	0.99	4.17	0.03	0.34	1.4	2.19	9.57	9.83
		Interval	40	100	1.24	1.59	0.62	3.96	0.025	0.30	0.89	1.37	5.40	7.61
Test House	Deposition Coupons	Bedroom	5	100	1.89	2.12	1.07	3.81	0.14	0.83	1.26	1.70	5.54	5.54
		Den	28	100	2.23	2.57	1.64	2.07	0.63	0.79	1.68	2.65	3.77	14.4
		Kitchen	24	100	31.6	56.4	11.5	4.16	1.0	5.00	9.12	25.2	179	229
		All	57	100	14.6	39.0	3.58	4.46	0.14	1.26	2.82	8.66	61.0	229
	Surface Wipe	Kitchen	9	100	1548	2793	627	3.71	120	270	470	1370	8880	8880

NC, Not calculated

LWW, Lioy-Weisel-Wainman sampler

Table A.21 Summary statistics for diazinon in Total Available Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
NHEXAS-AZ	Surface Wipe	Window Sill	6	0	NC	NC	NC	NC	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0
MNCPES	LWW	Floor	99	7	NC	NC	NC	NC	<3.5	<3.5	<3.5	<3.5	3.55	7.01
CCC	Surface Wipe	Floor	168	54	0.21	1.44	0.011	9.1	0.001	0.002	0.004	0.06	0.53	18.3
		Desk/Table	80	41	NC	NC	NC	NC	0.001	0.002	0.002	0.02	0.28	2.40
JAX	Surface Wipe	Floor (Screening)	46	89	1.35	5.07	0.11	10.5	<0.002	0.03	0.11	0.52	3.33	32.9
		Floor	9	44	NC	NC	NC	NC	<0.002	<0.002	<0.002	0.34	1.43	1.43
		Play Area	9	33	NC	NC	NC	NC	<0.002	<0.002	<0.002	0.002	3.99	3.99
CHAMACOS	Surface Wipe	All	20	95	0.041	0.033	0.024	3.73	<0.005	0.011	0.038	0.066	0.093	0.096
DIYC	Surface Wipe	Floor (Pre-application)	7	86	7.06	6.87	4.71	2.7	<0.3	2.61	3.85	10.3	20.8	20.8
		Floor	35	100	12.7	20.4	6.35	2.9	0.71	3.93	5.54	7.54	71.6	85.1

NC, Not calculated
LWW, Lioy-Weisel-Wainman sampler

Table A.22 Summary statistics for cis-permethrin in Total Available Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CCC	Surface Wipes	Floor	168	60	0.14	0.36	0.022	6.3	0.002	0.004	0.02	0.08	0.79	2.81
		Surfaces	80	44	1.55	10.5	0.015	8.4	<0.005	<0.005	<0.005	0.06	0.46	89.8
JAX	Surface Wipes	Floor (Screening)	46	87	8.46	15.5	0.93	19.9	<0.005	0.19	2.22	10.0	32.2	75.8
		Floor	9	78	8.56	16.4	0.35	28.3	<0.005	0.13	0.24	1.69	42.4	42.4
		Play Area	9	67	1.57	3.2	0.09	23.3	<0.005	<0.005	0.04	0.89	9.77	9.77
CHAMACOS	Surface Wipe	All	20	85	0.21	0.36	0.1	6.8	<0.005	0.053	0.10	0.21	1.1	1.7

Table A.23 Summary statistics for trans-permethrin in Total Available Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CCC	Surface Wipe	Floor	168	62	0.25	0.71	0.031	8.1	<0.005	<0.005	0.03	0.17	1.17	6.96
		Desk/Table	80	60	3.23	24.7	0.027	9.0	<0.005	<0.005	0.02	0.11	0.92	219
JAX	Surface Wipe	Floor (Screening)	46	89	10.2	19.4	1.18	19.3	<0.005	0.26	2.93	11.7	40.0	94.3
		Floor	9	78	12.9	24.9	0.44	34.1	<0.005	0.12	0.34	3.48	66.6	66.6
		Play Area	9	89	2.06	4.41	0.14	19.8	<0.005	0.02	0.05	1.45	13.6	13.6
CHAMACOS	Surface Wipe	All	20	95	0.43	0.77	0.18	5.1	<0.002	0.14	0.23	0.39	2.3	3.6

Table A.24 Summary statistics for cyfluthrin in Total Available Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CCC	Surface Wipe	Floor	168	7	NC	NC	NC	NC	<0.006	<0.006	<0.006	<0.006	0.4	6.87
		Desk/Table	80	1	NC	NC	NC	NC	<0.006	<0.006	<0.006	<0.006	<0.006	0.80
JAX	Surface Wipe	Floor (Screening)	46	20	NC	NC	NC	NC	<0.006	<0.006	<0.006	<0.006	4.33	13.8
		Floor	9	33	NC	NC	NC	NC	<0.006	<0.006	<0.006	0.04	10.1	10.1
		Play Area	9	11	NC	NC	NC	NC	<0.006	<0.006	<0.006	<0.006	3.45	3.45
CHAMACOS	Surface Wipe	All	20	5	NC	NC	NC	NC	<0.05	<0.05	<0.05	<0.05	<0.05	0.40

NC, Not calculated

Transferable Surface Residue Loadings

Table A.25 Summary statistics for chlorpyrifos in Transferable Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	C18 Press	Floor	102	8	NC	NC	NC	NC	<0.33	<0.33	<0.33	<0.33	0.44	63.5
		Surface	102	5	NC	NC	NC	NC	<0.33	<0.33	<0.33	<0.33	<0.33	0.70
CTEPP (NC)	Surface Wipe	Home Floor	28	89	0.02	0.05	0.0063	4.6	<0.0007	0.0031	0.0066	0.012	0.15	0.21
		Kitchen Counter	18	89	0.03	0.04	0.008	5.8	<0.0007	0.003	0.007	0.045	0.14	0.14
	PUF Roller	Home	18	94	0.01	0.023	0.005	4.5	<0.0004	0.0015	0.0035	0.009	0.072	0.072
CTEPP (OH)	Surface Wipe	Home Floor	21	86	0.19	0.84	0.0043	8.8	<0.0007	0.001	0.003	0.013	0.11	3.86
		Kitchen Counter	13	62	0.068	0.21	0.0025	10	<0.0007	<0.0007	0.001	0.006	0.76	0.76
	PUF Roller	Home	13	85	0.25	0.89	0.0026	11	<0.0004	0.001	0.002	0.004	3.22	3.22
CHAMACOS	C18 Press	All	20	0	NC	NC	NC	NC	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09
CPPAES	Surface Wipe	Floor	41	100	0.052	0.054	0.026	4.02	0.002	0.014	0.031	0.074	0.163	0.179
Test House	C18 Press	Den/Kitchen	16	94	1.02	2.06	0.26	5.13	<0.03	0.11	0.23	0.52	6.86	6.86
	PUF Roller	Den/Kitchen	6	100	0.030	0.059	0.007	5.97	0.001	0.002	0.0045	0.017	0.15	0.15

NC, Not calculated

Table A.26 Summary statistics for diazinon in Transferable Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	C18 Press	Floor	102	8	NC	NC	NC	NC	<0.14	<0.14	<0.14	<0.14	0.55	13.0
		Surface	102	8	NC	NC	NC	NC	<0.14	<0.14	<0.14	<0.14	1.13	2.68
CTEPP (NC)	Surface Wipe	Home Floor	28	68	0.056	0.19	0.002	8.4	<0.0007	<0.0007	0.001	0.003	0.51	0.91
		Kitchen Counter	18	61	0.063	0.21	0.003	8.8	<0.0007	<0.0007	0.002	0.008	0.87	0.87
	PUF Roller	Home	18	67	0.075	0.22	0.004	13	<0.0004	<0.0004	0.003	0.034	0.93	0.93
CTEPP (OH)	Surface Wipe	Home Floor	21	38	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	0.001	0.01	0.05
		Kitchen Counter	13	31	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	0.001	0.21	0.21
	PUF Roller	Home	13	54	0.01	0.03	0.001	1.71	<0.0004	<0.0004	0.001	0.002	0.11	0.11
CHAMACOS	C18 Press	All	20	0	NC	NC	NC	NC	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
DIYC	C18 Press	Floor	9	89	10.9	9.11	6.5	3.5	<1.2	1.24	3.78	11.7	23.9	23.9
		Counter	3	67	NC	NC	NC	NC	<1.2	NC	3.18	NC	NC	9.46
		Play Area	3	33	NC	NC	NC	NC	<1.2	NC	<1.2	NC	NC	3.89

NC, Not calculated

Table A.27 Summary statistics for *cis*-permethrin in Transferable Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Surface Wipe	Home Floor	28	93	0.161	0.263	0.034	8.6	<0.0007	0.0071	0.0443	0.192	0.832	0.874
		Kitchen Counter	18	83	3.05	11.7	0.044	24	<0.0007	0.0062	0.0596	0.361	50.1	50.1
	PUF Roller	Home	18	83	0.164	0.319	0.020	13	<0.0004	0.0038	0.0229	0.139	1.13	1.13
CTEPP (OH)	Surface Wipe	Home Floor	21	71	0.28	1.13	0.011	12	<0.0007	<0.0007	0.009	0.064	0.19	5.2
		Kitchen Counter	13	39	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	0.006	0.78	0.78
	PUF Roller	Home	13	69	0.035	0.08	0.004	9.3	<0.0004	<0.0004	0.004	0.012	0.29	0.29
CHAMACOS	C18 Press	All	20	0	NC	NC	NC	NC	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2

NC, Not calculated

Table A.28 Summary statistics for trans-permethrin in Transferable Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Surface Wipe	Home Floor	28	93	0.157	0.268	0.027	9.8	<0.0007	0.005	0.04	0.19	0.83	1.01
		Kitchen Counter	18	83	3.48	13.5	0.041	26	<0.0007	0.006	0.026	0.375	57.4	57.4
	PUF Roller	Home	18	83	0.18	0.34	0.018	14	<0.0004	0.003	0.02	0.17	1.16	1.16
CTEPP (OH)	Surface Wipe	Home Floor	21	71	0.28	1.12	0.011	13	<0.0007	<0.0007	0.01	0.07	0.2	5.18
		Kitchen Counter	13	39	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	0.005	0.79	0.79
	PUF Roller	Home	13	69	0.03	0.08	0.003	8.2	<0.0004	<0.0004	0.003	0.008	0.29	0.29
CHAMACOS	C18 Press	All	20	0	NC	NC	NC	NC	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2

NC, Not calculated

Table A.29 Summary statistics for cyfluthrin using in Transferable Residue (ng/cm²).

Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Surface Wipe	Home Floor	28	7	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	<0.0007	0.05	0.13
		Kitchen Counter	18	0	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	<0.0007	<0.0007	<0.0007
	PUF Roller	Home	18	78	0.11	0.10	0.05	5.4	<0.0004	0.02	0.10	0.16	0.41	0.41
CTEPP (OH)	Surface Wipe	Home Floor	21	10	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	<0.0007	0.041	0.078
		Kitchen Counter	13	0	NC	NC	NC	NC	<0.0007	<0.0007	<0.0007	<0.0007	<0.0007	<0.0007
	PUF Roller	Home	13	0	NC	NC	NC	NC	<0.0004	<0.0004	<0.0004	<0.0004	<0.0004	<0.0004
CHAMACOS	C18 Press	All	20	0	NC	NC	NC	NC	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2

NC, Not calculated

Solid Food Concentrations and Intakes

Table A.30 Summary statistics for chlorpyrifos measured in solid food, presented as both intake (μg/day) and concentration (μg/kg).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Intake (μg/day)	MNCPES	Dup Diet	All	96	91	0.42	0.64	0.24	2.9	<0.12	0.14	0.26	0.38	1.6	4.8
	CTEPP-NC	Dup Diet/ Dup Plate	All	129	75	0.20	0.66	0.079	3.3	<0.024	0.029	0.093	0.18	0.64	7.3
	CTEPP-OH	Dup Diet/ Dup Plate	All	125	78	0.13	0.18	0.073	2.7	<0.024	0.035	0.071	0.13	0.40	1.3
	JAX	Dup Diet	All	9	100	1.3	1.6	0.76	3.0	0.12	0.48	1.1	1.2	5.2	5.2
Concentration (μg/kg)	NHEXAS-AZ	Dup Diet	≤2 years	20	15	NC	NC	NC	NC	<1.0	<1.0	<1.0	<1.0	5.7	7.2
	MNCPES	Dup Diet	All	96	88	0.79	1.2	0.51	2.3	<0.26	0.29	0.53	0.81	2.4	7.1
	CTEPP (NC)	Dup Diet/ Dup Plate	Home	129	65	0.57	1.8	0.20	3.4	<0.08	<0.08	0.19	0.39	2.1	20
			Daycare	24	54	0.23	0.25	0.14	2.7	<0.08	<0.08	0.10	0.35	0.85	0.95
	CTEPP (OH)	Dup Diet/ Dup Plate	Home	125	66	0.38	0.61	0.19	3.0	<0.08	<0.08	0.19	0.39	1.6	3.5
			Daycare	29	69	0.20	0.19	0.15	2.3	<0.08	<0.08	0.14	0.24	0.56	0.88
	JAX	Dup Diet	All	9	100	1.3	2.3	0.51	4.2	0.050	0.25	0.38	1.5	7.4	7.4
	CHAMACOS	Dup Diet	All	17	6	NC	NC	NC	NC	<1.0	<1.0	<1.0	<1.0	1.4	1.4

Dup Diet, Duplicate Diet; Dup Plate, Duplicate Plate
NC, Not calculated

Table A.31 Summary statistics for diazinon measured in solid food, presented as both intake (μg/day) and concentration (μg/kg).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Intake (μg/day)	MNCPES	Dup Diet	All	101	20	NC	NC	NC	NC	<0.019	<0.019	<0.019	<0.019	0.12	0.64
	CTEPP-NC	Dup Diet/ Dup Plate	All	128	32	NC	NC	NC	NC	<0.024	<0.024	<0.024	0.040	0.095	1.3
	CTEPP-OH	Dup Diet/ Dup Plate	All	125	23	NC	NC	NC	NC	<0.024	<0.024	<0.024	<0.024	0.073	0.21
	DIYC	Dup Diet	All	16	100	0.42	0.29	0.34	2.0	0.095	0.23	0.30	0.51	1.1	1.1
	JAX	Dup Diet	All	9	11	NC	NC	NC	NC	<0.35	<0.35	<0.35	<0.35	0.67	0.67
Concentration (μg/kg)	NHEXAS-AZ	Dup Diet	≤12 years	20	10	NC	NC	NC	NC	<0.7	<0.7	<0.7	<0.7	1.8	1.9
	MNCPES	Dup Diet	All	101	6	NC	NC	NC	NC	<0.2	<0.2	<0.2	<0.2	0.22	2.0
	CTEPP (NC)	Dup Diet/ Dup Plate	Home	128	22	NC	NC	NC	NC	<0.08	<0.08	<0.08	<0.08	0.41	6.7
			Daycare	24	25	NC	NC	NC	NC	<0.08	<0.08	<0.08	0.08	0.17	0.89
	CTEPP (OH)	Dup Diet/ Dup Plate	Home	125	15	NC	NC	NC	NC	<0.08	<0.08	<0.08	<0.08	0.18	0.72
			Daycare	29	24	NC	NC	NC	NC	<0.08	<0.08	<0.08	<0.08	0.20	0.23
	DIYC	Dup Diet	All	16	100	0.29	0.25	0.23	1.9	0.12	0.15	0.17	0.31	1.01	1.01
	JAX	Dup Diet	All	9	44	NC	NC	NC	NC	<0.04	<0.04	<0.04	0.080	1.05	1.05
	CHAMACOS	Dup Diet	All	17	12	NC	NC	NC	NC	<1	<1	<1	<1	1.0	1.0

Dup Diet, Duplicate Diet; Dup Plate, Duplicate Plate
NC, Not calculated

Table A.32 Summary statistics for *cis*-permethrin measured in solid food, presented as both intake (μg/day) and concentration (μg/kg).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Intake (μg/day)	MNCPES	Dup Diet	All	100	30	NC	NC	NC	NC	<0.019	<0.019	<0.019	<0.019	0.92	2.6
	CTEPP-NC	Dup Diet	All	129	50	2.7	14	0.10	7.3	<0.024	<0.024	0.060	0.23	6.8	93
	CTEPP-OH	Dup Diet	All	125	38	NC	NC	NC	NC	<0.024	<0.024	<0.024	0.090	4.8	113
Concentration (μg/kg)	MNCPES	Dup Diet	All	100	20	NC	NC	NC	NC	<0.024	<0.024	<0.024	0.14	1.5	4.9
	CTEPP (NC)	Dup Diet/ Dup Plate	Home	129	46	NC	NC	NC	NC	<0.08	<0.08	<0.08	0.59	16	81
			Daycare	24	25	NC	NC	NC	NC	<0.08	<0.08	<0.08	0.22	5.2	218
	CTEPP (OH)	Dup Diet/ Dup Plate	Home	125	31	NC	NC	NC	NC	<0.08	<0.08	<0.08	0.19	8.8	560
			Daycare	29	24	NC	NC	NC	NC	<0.08	<0.08	<0.08	<0.08	2.2	31
	JAX	Dup Diet	All	9	78	1.6	4.2	0.19	7.9	<0.02	0.080	0.29	0.35	13	13

Dup Diet, Duplicate Diet; Dup Plate, Duplicate Plate
NC, Not calculated

Table A.33 Summary statistics for *trans*-permethrin measured in solid food, presented as both intake (μg/day) and concentration (μg/kg).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Intake (μg/day)	MNCPES	Dup Diet	All	101	13	NC	NC	NC	NC	<0.01	<0.01	<0.01	<0.01	0.15	1.4
	CTEPP-NC	Dup Diet/ Dup Plate	All	128	50	1.5	8.0	0.087	6.1	<0.024	<0.024	0.051	0.19	4.6	65
	CTEPP-OH	Dup Diet/ Dup Plate	All	125	38	NC	NC	NC	NC	<0.024	<0.024	<0.024	0.069	4.2	90
Concentration (μg/kg)	MNCPES	Dup Diet	All	101	7	NC	NC	NC	NC	<0.08	<0.08	<0.08	<0.08	0.33	1.9
	CTEPP (NC)	Dup Diet/ Dup Plate	Home	128	46	NC	NC	NC	NC	<0.08	<0.08	<0.08	0.58	8.7	70
			Daycare	24	25	NC	NC	NC	NC	<0.08	<0.08	<0.08	0.18	3.0	149
	CTEPP (OH)	Dup Diet/ Dup Plate	Home	125	31	NC	NC	NC	NC	<0.08	<0.08	<0.08	0.18	8.0	448
			Daycare	29	24	NC	NC	NC	NC	<0.08	<0.08	<0.08	<0.08	1.4	27
	JAX	Dup Diet	All	9	78	2.8	7.3	0.27	9.8	<0.02	0.17	0.22	0.45	22	22

Dup Diet, Duplicate Diet; Dup Plate, Duplicate Plate
NC, Not calculated

Table A.34 Summary statistics for TCPy measured in solid food, presented as both intake (μg/day) and concentration (μg/kg).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Intake (μg/day)	CTEPP-NC	Dup Diet/ Dup Plate	All	128	99	1.4	0.97	0.99	2.6	<0.038	0.71	1.2	1.8	3.4	5.5
	CTEPP-OH	Dup Diet/ Dup Plate	All	127	100	1.0	0.90	0.70	2.5	0.038	0.41	0.77	1.4	2.3	7.8
Concentration (μg/kg)	CTEPP (NC)	Dup Diet/ Dup Plate	Home	128	98	3.1	2.8	2.1	2.6	<0.12	1.5	2.3	3.8	8.6	18
			Daycare	24	100	3.8	3.3	2.8	2.3	0.25	2.3	2.9	4.5	6.6	18
	CTEPP (OH)	Dup Diet/ Dup Plate	Home	127	99	2.6	2.6	1.7	2.7	<0.13	1.0	1.9	3.3	5.8	23
			Daycare	29	100	2.8	5.0	1.7	2.4	0.38	0.98	1.5	2.5	8.1	27
	JAX	Dup Diet	All	9	100	5.0	3.7	4.0	1.9	2.0	2.4	3.2	7.1	12	12

Dup Diet, Duplicate Diet; Dup Plate, Duplicate Plate
NC, Not calculated

Table A.35 Summary statistics for IMP measured in solid food, presented as both intake (μg/day) and concentration (μg/kg).

	Study	Method	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
Intake (μg/day)	CTEPP-OH	Dup Diet/ Dup Plate	All	32	97	0.19	0.17	0.14	2.2	<0.024	0.093	0.12	0.20	0.58	0.63
Concentration (μg/kg)	CTEPP (OH)	Dup Diet/ Dup Plate	Home	40	88	0.52	0.54	0.36	2.4	<0.12	0.26	0.33	0.63	1.6	2.7
			Daycare	29	83	0.40	0.29	0.30	2.3	<0.13	0.14	0.35	0.58	0.90	1.2

Dup Diet, Duplicate Diet; Dup Plate, Duplicate Plate
NC, Not calculated

Hand Loadings

Table A.36 Summary statistics for chlorpyrifos hand loadings (ng/cm^2).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
MNCPES	Rinse	97	39	NC	NC	NC	NC	<0.07	<0.07	<0.07	0.094	0.27	3.1
CTEPP (NC)	Home	96	78	0.053	0.11	0.020	3.9	<0.007	0.0082	0.020	0.046	0.28	0.74
	Daycare	31	68	0.023	0.022	0.013	3.4	<0.007	<0.007	0.017	0.036	0.073	0.077
CTEPP (OH)	Home	97	55	0.18	1.5	0.011	4.8	<0.007	<0.007	0.011	0.029	0.17	15
	Daycare	29	55	0.036	0.11	0.010	4.0	<0.007	<0.007	0.010	0.021	0.075	0.58
CPPAES	Rinse	38	100	2.8	3.1	1.6	3.2	0.09	0.74	1.9	3.7	11	18
	Wipe	44	100	0.32	0.29	0.19	3.3	0.016	0.87	0.30	0.42	0.77	1.5

NC, Not calculated

Table A.37 Summary statistics for diazinon hand loadings (ng/cm^2).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Home	96	36	NC	NC	NC	NC	<0.005	<0.005	<0.005	0.011	0.084	1.6
	Daycare	31	55	0.015	0.032	0.0069	3.0	<0.005	<0.005	0.0065	0.014	0.051	0.17
CTEPP (OH)	Home	97	31	NC	NC	NC	NC	<0.005	<0.005	<0.005	0.0068	0.075	3.8
	Daycare	29	31	NC	NC	NC	NC	<0.005	<0.005	<0.005	0.0071	0.043	0.093
PET	Feasibility	15	100	0.32	0.29	0.19	3.6	<0.005	<0.005	<0.005	<0.005	0.94	0.94
DIYC	All	13	100	0.12	0.063	0.092	2.3	<0.005	<0.005	<0.005	<0.005	0.21	0.21

NC, Not calculated

Table A.38 Summary statistics for *cis*-permethrin hand loadings (ng/cm^2).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Home	96	86	0.92	6.5	0.071	6.7	<0.005	0.026	0.062	0.26	1.5	64
	Daycare	31	94	0.17	0.38	0.067	3.9	<0.005	0.035	0.073	0.15	0.31	2.2
CTEPP (OH)	Home	97	88	0.14	0.30	0.039	4.9	<0.005	0.017	0.033	0.095	0.88	2.1
	Daycare	29	79	0.15	0.29	0.034	6.5	<0.005	0.010	0.035	0.14	0.65	1.4

NC, Not calculated

Table A.39 Summary statistics for *trans*-permethrin hand loadings (ng/cm^2).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Home	96	86	0.93	6.8	0.055	6.9	<0.005	0.015	0.049	0.18	1.3	67
	Daycare	31	94	0.14	0.38	0.046	4.0	<0.005	0.020	0.036	0.12	0.26	2.1
CTEPP (OH)	Home	97	88	0.13	0.34	0.032	4.9	<0.005	0.013	0.027	0.072	0.77	2.1
	Daycare	29	79	0.15	0.33	0.030	6.5	<0.005	0.011	0.028	0.087	0.83	1.5

NC, Not calculated

Table A.40 Summary statistics for TCPy hand loadings (ng/cm^2).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (NC)	Home	99	100	0.023	0.022	0.018	1.9	0.0041	0.012	0.019	0.026	0.054	0.17
	Daycare	32	94	0.012	0.0076	0.010	2.0	<0.003	0.0066	0.010	0.017	0.029	0.032
CTEPP (OH)	Home	98	98	0.015	0.012	0.012	2.0	<0.003	0.0079	0.012	0.019	0.033	0.067
	Daycare	29	90	0.012	0.0075	0.010	1.9	<0.003	0.0062	0.011	0.015	0.030	0.036

NC, Not calculated

Table A.41 Summary statistics for IMP hand loadings (ng/cm^2).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP (OH)	Home	98	49	NC	NC	NC	NC	<0.003	<0.003	<0.003	0.0040	0.017	0.064
	Daycare	29	31	NC	NC	NC	NC	<0.003	<0.003	<0.003	0.0031	0.017	0.043

NC, Not calculated

Urinary Metabolite Concentrations

Table A.42 Summary statistics for TCPy measured in urine (ng/mL).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
NHEXAS-AZ	≤2 years	21	100	12	7.6	9.3	2.2	2.0	5.7	12	14	26	30
MNCPES	All	263	92	9.2	7.7	6.6	2.3	<1.4	4.0	7.2	12	23	45
CTEPP-NC	All	129	98	7.5	10	5.5	2.1	<1.0	3.8	5.3	8.4	16	100
CTEPP-OH	All	123	100	5.9	3.5	4.9	1.9	1.2	3.1	5.2	7.8	12	15
JAX	All	9	100	11	6.4	9.1	2.1	2.9	7.5	9.8	15	21	21
CPPAES	All	81	93	8.0	4.7	6.4	2.1	<1.0	4.5	7.7	11	18	20
NHANES	≤2 years	1245	90	4.7	6.1	2.6	3.2	<0.4	1.3	2.8	6.0	15	64

Table A.43 Summary statistics for 3-PBA measured in urine (ng/mL).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
CTEPP-OH	All	126	68	0.81	3.0	0.38	2.6	<0.20	<0.20	0.32	0.69	1.9	34
JAX	All	9	100	19.6	33	3.9	7.5	0.39	0.76	2.2	29	99	99
NHANES	≤2 years	679	79	1.4	10	0.36	3.7	<0.10	0.13	0.34	0.78	3.8	254

Table A.44 Summary statistics for IMP measured in urine (ng/mL).

Study	Group	n	%Det	Mean	SD	GM	GSD	Min	25th	50th	75th	95th	Max
PET	All	30	77	1.3	1.6	0.75	2.8	<0.22	0.39	0.62	1.5	5.5	6.2
DIYC	All	41	100	9.0	6.9	7.1	2.0	1.7	4.4	7.1	10	27	29
NHANES	≤2 years	1220	15	NC	NC	NC	NC	<0.7	<0.7	<0.7	<0.7	3.0	145

NC, Not calculated

APPENDIX B: Individual Study Details

National Human Exposure Assessment Survey in Arizona (NHEXAS-AZ)

Collaborators: University of Arizona, Battelle Memorial Institute, and the Illinois Institute of Technology

Study Design:

- Type: Observational exposure measurement study with probability-based sample
- Location: Each of the 15 counties in Arizona
- Monitoring period: December 1995 to March 1997
- Study population: 176 households (this report only includes data from 21 households in which the primary participants were children, ages 6-12)
- Pesticide Use: Participants did not report use prior to the study

Monitoring Protocol:

- Indoor and Outdoor air: 3-day integrated samples; Personal air: 1-day sample
- Surface Dust Loading: Modified Hoover "Port-a-Power" vacuum, center and corner of living room and bedroom; Window sill wipes
- Soil: Yard surface soil composite sample
- Beverages and solid food: 24-hour duplicate diet
- Hand wipes: 4-mL IPA wipes of both hands
- Urine: First morning void samples
- Activities: Baseline and follow-up questionnaires, time-activity diary
- Analytes (Pesticides):
    - Two pesticides of primary interest (and metabolites), namely chlorpyrifos (TCPy) and diazinon, and 14 secondary pesticides, including malathion (MDA) and carbaryl (1-naphthol)

Key Outputs:

- Occurrence, distributions, and determinants of total exposure to the general population
- Geographic trends in multimedia exposure
- Total exposures in minority and disadvantaged subsets of the population

Minnesota Children's Pesticide Exposure Study (MNCPES)

Collaborators: RTI, EOHSI, University of Minnesota, and Minnesota Department of Health

Study Design:

- Type: Observational exposure measurement study with probability-based sample
- Location: Minneapolis/St. Paul (urban) and Goodhue and Rice counties (rural)
- Monitoring period: Summer 1997
- Study population: 102 children, ages 3-13
- Pesticide Use: Households reporting a history of more frequent pesticide use were oversampled

Monitoring Protocol:

- Environmental samples:
  - Personal, indoor, and outdoor air: Integrated samples, days 1-7 (outdoor air for only 10% of urban homes)
  - Surface dust loading: Wipe and press, 2 indoor locations (main play area and family room), day 4
  - Soil: Surface soil grab sample, day 4
  - Beverages and solid food: Duplicate diet, 4-d composite, days 3-6
  - Tap water: Grab sample (10% urban homes), day 4
- Biological/Personal samples:
  - Hand rinse, day 3
  - Urine: First morning void samples (88%) 3 samples per child, days 3, 5, and 7
- Activities:
  - Baseline and follow-up questionnaires, time-activity diary
  - Videotape (4-h, about 20 homes)
- Analytes (Pesticides and PAHs):
  - Pesticides: 4 Primary pesticides and metabolites, namely chlorpyrifos (TCPy), atrazine (atrazine mercapturate), malathion (malathion dicarboxylic acid), and diazinon, and 14 secondary pesticides
  - PAHs: 13 PAHs including fluoranthene, phenanthrene, and pyrene

Key Outputs:

- An "inverse" PK model to predict chlorpyrifos dose resulting both from specific pesticide applications and from average low-level exposures
- Distributions and correlations in environmental and biological media (Adgate *et al.,* 2001; Clayton *et al.,* 2003)
- Evaluation of pathways of exposure

Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants Study (CTEPP)

Collaborators: Battelle

Study Design:

- Type: Observational exposure measurement study with probability-based sample in homes and child care centers
- Location: North Carolina (NC) and Ohio (OH)
- Monitoring period: NC (July 2000 to March 2001); OH (April 2001 to November 2001)
- Study population: 257 children, ages 18 months to five years, and their primary adult caregivers (NC = 130 children, 130 homes, 13 daycare centers; OH = 127 children; 127 homes, 16 daycare centers)
- Pesticide use: Use during previous seven days were reported by a subset (n=38) of families in their homes

Monitoring Protocol:

- Sampling times: Samples collected over a 48-hr period at a home and/or daycare center:
- Samples/data collected: Soil, outdoor air, indoor air, indoor floor dust, hand wipe, liquid food, solid food, urine
- Supplemental information:
  - Recruitment survey, house/building characteristics survey, pre- and post monitoring questionnaires, activity and food diaries
  - In addition, 20% of the participants from OH were videotaped about 2 hours at their homes
  - Additional samples were collected if a pesticide was reported by the participant as having been applied indoors or outdoors at a home or daycare center within 7 days of previously scheduled field sampling or during the 48-hr monitoring period (hard floor surface wipe, food preparation surface wipe, and transferable residue)
- Analytes of interest: Chlorpyrifos, diazinon, and *cis*-/*trans*-permethrin

Key Outputs:

- Pesticide distributions in microenvironments where children spend time
- Transfer of pesticides from microenvironmental media to child and factors that affect transfer
- Evaluation of pathways of exposure
- Evaluation of important factors the affect exposure

First National Environmental Health Survey of Child Care Centers (CCC)

Collaborators: HUD, CPSC (US Department of Housing and Urban Development, US Consumer Product Safety Commission)

Study Design:

- Type: Observational study with probability-based sample of licensed child care centers
- Location: Nationwide
- Monitoring period: August 2001 to October 2001
- Study population: 168 child care centers; no children or adults participated in the study
- Pesticide use: Child care center directors reported on the professional or center staff applications during the previous 12 months

Monitoring Protocol:

- One time visit by field technicians to each child care center
- Samples collected: Soil, surface wipes, transferable residues (surface press)
- Analytes: Current-use pesticides – organophosphates and pyrethroids

Key Outputs:

- Data relating to pesticide use practices in child care centers across the US
- Characterization of spatial distribution and magnitude of pesticide concentrations on surfaces in a sample of U.S. child care centers

Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposures in Children Living in Jacksonville, Florida (JAX)

Collaborators: CDC (Centers for Disease Control and Prevention), DCHD (Duval County Health Department)

Study Design:

- Type: Observational pilot exposure measurement study
- Location: Jacksonville, Florida (Duval County)
- Monitoring period: August to October 2001
- Study population: Nine children 4-6 years of age
- Pesticide use: Participants report recent pesticide use in the residences

Monitoring Protocol:

- Sampling times: One-time sample collection with 24-hour air samples
- Samples collected:
    - Surface wipe
    - Indoor/outdoor air
    - Duplicate diet
    - Transferable residues
    - Cotton garments
    - Urine
- Questionnaires:
    - Pesticide screening inventory
    - Time activity diary
- Analytes: OP, pyrethroid pesticides, metabolites in urine

Key Outputs:

- The CDC component of the study determined the distribution of urinary metabolite levels of organophosphate and pyrethroid pesticides in a group of 4-6 year old children living in the greater Jacksonville, Florida area
- The DCHD component of the study evaluated the use of screening wipes and pesticide inventories to identify homes with potentially elevated pesticide levels and to identify potential household sources for pesticides
- The EPA nine-home study was performed to evaluate methods for aggregate exposure measurements, to determine whether environmental measures of pesticide exposure are correlated with biological samples for a sub-sample of homes using pesticide inventories and screening measurements, to evaluate if information collected from pesticide screening inventories about pesticides used in the home correlates with environmental measures found in the same homes, and to evaluate pathways of exposure and the important factors that affect exposure

Center for the Health Assessment of Mothers and Children of Salinas Quantitative Exposure Assessment Study (CHAMACOS)

Collaborator: University of California at Berkeley

Study Design:

- Type: Observational pilot exposure measurement study
- Location: Salinas, California
- Monitoring period: June 2002 to October 2002
- Study population: Twenty children ages 5 to 35 months old, 10 female, 10 male
- Pesticide use: Incidental for farmworker children

Monitoring Protocol:

- Sampling times: 24-hour monitoring
- Samples collected:
    - Indoor and outdoor air
    - House dust
    - Transferable residues from floors (surface wipes and press samples)
    - Transferable residues from toys (surface wipes)
    - Cotton union suits and socks
    - Urine
- Activities
    - Videotaping
    - Time-activity diary
- Analytes: acephate, azinphos methyl, bifenthrin, chlorpyrifos, chlorpyrifos oxon, *cis*-allethrin, *trans*-allethrin, *cis*-permethrin, *trans*-permethrin, cyfluthrin (I, II, III, IV), cypermethrin (I, II, II, IV), dacthal, deltamethrin (I, II), diazinon, dimethoate, esfenvalerate, fonofos, iprodione, *lambda*-cyhalothrin, malathion, methidathion, naled, p,p'-DDE, p,p'-DDT, phosmet, resmethrin, sumithrin, tetramethrin (I, II), vinclozilline

Key Outputs:

- Evaluation of methods for aggregate exposure measurements
- Pesticide distributions in microenvironments where children spend time
- Transfer of pesticides from microenvironmental media to child and factors that affect transfer
- Evaluation of pathways of exposure and important factors that affect exposure

Children's Pesticide Post-Application Exposure Study (CPPAES)

Collaborator: EOHSI (Environmental and Occupational Health Sciences Institute)

Study Design:

- Type: Observational pilot exposure measurement study
- Location: Urban New Jersey
- Monitoring period: April 1999 to March 2001
- Study population: 10 homes; children 2-5 years of age
- Pesticide use: Crack and crevice application of chlorpyrifos was applied by a professional applicator in these homes

Monitoring Protocol:

- Sampling times: 1 day prior to application, 1, 2, 3, 5, 7, 9, and 11 days after application
- Samples collected:
    - All sampling days: indoor air, deposition coupons, surface samples (LWW), toys, hand wipes, urine, air exchange rate, time activity diary
    - Additional day 2 samples - surface wipes, hand wipes, dermal wipes, cotton garments, videotaping
- Analyte: Chlorpyrifos, TCPy in urine

Key Output:

- Pesticides distributions in microenvironments where children spend time
- Transfer of pesticide from microenvironmental media to child and factors that affect transfer
- Evaluation of pathways of exposure
- Evaluation of important factors that affect exposure

The Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the US EPA Indoor Air Quality Research Test House (Test House)

Collaborator: National Risk Management Research Laboratory

Study Design:

- Type: Field laboratory (Indoor Air Quality Research Test House)
- Location: Cary, NC
- Monitoring period: 3 weeks during November 2000
- Study population: Single residential house; no occupants in the test house
- Pesticide use: Chlorpyrifos, EC formulation, crack and crevice application in kitchen area (floor and cabinetry)

Monitoring Protocol:

- Sampling intervals: Pre, 1, 3, 7, 14 and 21 days post application
- Sample types:
    - Application formulation concentration
    - Air (kitchen, den and master bedroom)
    - PUF-skin roller (den and kitchen)
    - Carpet sections (den and master bedroom)
    - 10-min C18 surface press (den carpet and kitchen vinyl floor), wipes (kitchen floor and counter)
- Analyte: Chlorpyrifos

Key Outputs:

- Translocation and exposure pathways
- Inputs to algorithms and SHEDS
- Temporal and spatial variability over sampling period

A Pilot Study Examining Translocation Pathways Following a Granular Application of Diazinon to Residential Lawns (PET)

Collaborators: None

Study Design:

- Preceded by a 1-home feasibility study
- Type: Observational pilot exposure measurement study residential homes
- Location: 50 mile radius of Durham, NC
- Monitoring period: Ten days in Spring 2001
- Study population: 6 homes, 1 child and care giver (typically mother)
- Pesticide use: Homeowner applied diazinon, granular formulation, residential lawns (turf)

Monitoring Protocol:

- Sampling intervals: Pre, 1, 2, 4 and 8 days post application
- Sample types:
    - Application formulation concentration
    - Air (living room and child's bedroom)
    - PUF roller (lawn and indoor floor)
    - Soil
    - Entryway doormat
    - HVS3
    - Cotton gloves (technician and child)
    - Urine (adult and child)
    - Dog fur clippings
    - Dog paw wipes
    - Dog blood
    - Videography (15-min)

Key Outputs:

- Methods evaluation
- Translocation and exposure pathways
- Decay rates over sampling period
- Inputs to algorithms and SHEDS

Dietary Intake of Young Children (DIYC)

Collaborator: RTI

Study Design:

- Type: Observational pilot exposure measurement study
- Location: Raleigh, NC area
- Monitoring period: November 1999 to January 2000
- Study population: 3 homes; children 1-3 years old
- Pesticide use: Diazinon applications reported by homeowner - commercial crack and crevice (2 homes) or applied by resident (1 home)

Monitoring Protocol:

- Sampling times: Pre-application to 8 days post-application (7 visits total)
- Samples collected:
    - Indoor and outdoor air
    - Surface wipes (isopropanol)
    - Entry wipe
    - PUF roller
    - Surface press
    - Hand wipes
    - Food press
    - Food samples
    - Urine
- Analyte: Diazinon

Key Outputs:

- Evaluation of methods to measure excess dietary exposures that occur from activities by young children during eating
- Children's dietary intake model accurately represents total dietary exposures of children
- Model predictions are closest to measured results with the highest measured environmental diazinon concentrations
- Refinements for transfer and activity parameters within model are needed
- Categories of transfers and activities for highly exposed vs. less exposed are needed

Characterizing Pesticide Residue Transfer Efficiencies (Transfer)

Collaborator: Battelle

Study Design:

- Type: Controlled laboratory study
- Objective: Evaluate parameters that affect residue transfer from surface-to-skin, skin-to-other objects, skin-to-mouth, and object-to-mouth
- Monitoring period: not applicable
- Study population: not applicable
- Pesticide use: Nontoxic fluorescent tracers used as surrogates for pesticides

Monitoring Protocol:

- Conduct study using nontoxic fluorescent tracers as a surrogate for pesticide residues
- Apply fluorescent tracer as a residue at levels typical of residential pesticide applications to surfaces of interest
- Conduct controlled transfer experiments varying parameters in a systematic fashion
- Hand Contact Trials
    - Systematically varied six parameters
    - Repetitive contacts with contaminated surface
- Transfer off skin
    - Hand to clean surface
    - Hand to washing solution
    - Hand to mouth
- Mouthing Trials
    - Varied 5 parameters
    - Simulated mouthing using saliva moistened PUF
    - Measured mass of tracer transferred and estimated contact surface area using video imaging techniques
- Conduct laboratory evaluations to relate transfer of tracer to transfer of pesticides

Key Outputs:

- Transfer efficiency data
- Information on type of microactivity data needed to estimate dermal exposure
- Inputs for multipathway exposure models

Feasibility of Macroactivity Approach to Assess Dermal Exposure (Daycare)

Collaborator: RTI

Study Design:

- Type: Observational pilot exposure measurement study
- Location: North Carolina
- Monitoring period: Three occasions, twice per occasion
- Study population: Infants and toddlers at daycare centers
- Pesticide use: Professional crack and crevice applications as contracted by the daycare center

Monitoring Protocol:

- Identify up to 9 daycare centers with previously established contracts for routine monthly pesticide applications
- In each daycare, conduct screening sampling to evaluate the distribution of transferable pesticide residue on floor surfaces in the area where children spend the most time
- Select one daycare for intensive measurements
- Children from different age groups volunteered to wear full-body cotton bodysuits for short time periods
- Conduct surface sampling and videotaping of activities simultaneously with dermal loading sampling
- Calculate dermal transfer coefficients

Key Outputs:

- Pesticide distributions in nine daycare centers
- Verified protocol for collecting aggregate surface measurement
- Verified protocol for collecting transfer coefficients
- Dermal transfer coefficients developed with children (to evaluate default assumptions used in OPP's SOPs)

Food Transfer Studies, also known as Press Evaluation Studies (Food)

Collaborator: RTI

Study Design:

- Type: Controlled laboratory study
- Location: NERL Cincinnati
- Study period: Not applicable
- Study population: Not applicable
- Pesticide use: Organophosphate, pyrethroid, and pyrazole insecticides on various household surfaces

Monitoring Protocol:

- Surfaces:
    - Surface Treatment: A customized spray chamber was used to spray Pesticide Spray Solution (PSS) onto the ceramic tiles
    - Surface Drying: Following spraying, each ceramic tile was transferred to a glove box where it was air dried for an hour at constant temperature and humidity
    - Surface Wipes: Pesticide transfer to foods were compared to the pesticides removed using surface wipes (isopropanol moistened gauze pads), which were wiped across the ceramic tile in both the horizontal and vertical direction
- Food Items:
    - Moisture Content: Moisture (%) content measured with a Denver Instrument IR-30 moisture meter
    - Fat Content: Fate (%) content determined from each food's Nutrition Facts label; % fat = [total fat (g) / food serving size (g)] *100
    - Food Items: Pesticide transfer efficiencies were measured for three different foods, with standardized surface contact area; the foods were Fruit Roll-Ups Blastin' Berry Hot Colors® (Betty Crocker®), thinly sliced bologna (made with chicken & pork), and Red Delicious apple slices
- Transfer Efficiency (TE): TE is defined as the amount of pesticide recovered from the food item divided by the pesticide concentration or loading level
- Analytes: Malathion, Chlorpyrifos, Fipronil, Permethrin, Cyfluthrin, Cypermethrin, Deltamethrin

Key Outputs:

- Determine the extent of pesticide transfer from household surfaces to foods
- Evaluate factors that have been identified as important, including surface type, duration of contact, surface loading, and contact pressure (applied force or weight per area)
- Compared surface wipes using cotton gauze pads with the pesticide transfer to the foods

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